Vol. I

TRANSCRIPT OF RECORD

Supreme Court of the United States

OCTOBER TERM, 1941

No. 680

U. S. INDUSTRIAL CHEMICALS, INC., PETITIONER,

vs.

CARBIDE AND CARBON CHEMICALS CORPORATION

ON WRIT OF CERTIORARI TO THE UNITED STATES CIRCUIT COURT OF APPEALS FOR THE FOURTH CIRCUIT.

PETITION FOR CERTIORARI FILED OCTOBER 4, 1941.

CERTIORARI GRANTED NOVEMBER 10, 1941.

United States Circuit Court of Appeals

FOR THE FOURTH CIRCUIT

No. 4774

U. S. INDUSTRIAL CHEMICALS, INC.,
Defendant-Appellant,

v.

CARBIDE AND CARBON CHEMICALS CORPORATION,
Plaintiff-Appellee.

APPEAL FROM THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF MARYLAND

SUPPLEMENT TO DEFENDANT-APPELLANT'S BRIEF

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SUPPLEMENT TO DEFENDANT-APPELLANT'S BRIEF

District Court of the United States

FOR THE DISTRICT OF MARYLAND

CARBIDE AND CARBON CHEMICALS CORPORATION, *Plaintiff*, v. U. S. INDUSTRIAL CHEMICALS, INC., *Defendant*.	CIVIL ACTION No. 249

INTERLOCUTORY DECREE

This cause came on to be heard on the pleadings and proofs offered by all parties, and was argued by counsel, and thereupon, upon consideration thereof, it was ORDERED, ADJUDGED and DECREED in accordance with the opinion herein rendered:

1. That United States Reissue Letters Patent No. 20,370 granted May 18, 1937 to plaintiff, Carbide and Carbon Chemicals Corporation, a corporation of New York, as assignee of Theodore Emile Lefort, for process for the production of ethylene oxide, were issued upon an application filed September 25, 1936 for reissue of United States Letters Patent No. 1,998,878 which had been issued April 23, 1935;

2. That said United States Reissue Letters Patent No. 20,370 are good and valid in law and in equity as to claims 8 and 9 thereof;

3. That said Theodore Emile Lefort was the first original and sole inventor of the subject matter described in said Reissue Letters Patent No. 20,370 and claimed in claims 8 and 9 thereof;

4. That plaintiff, Carbide and Carbon Chemicals Corporation, is and has been since the grant of said reissue letters patent, possessed of the full and entire right, title and interest in and to the same;

5. That defendant, U. S. Industrial Chemicals, Inc., a corporation of Delaware, maintains a regular and established place of business within the District of Maryland and has there infringed upon claims 8 and 9 of the said Reissue Letters Patent No. 20,370 by practicing a process for the production of ethylene oxide, as set forth in Plaintiff's Exhibit 7

6. That plaintiff, Carbide and Carbon Chemicals Corporation, recover from the defendant, U. S. Industrial Chemicals, Inc., the profits, gains and advantages which it, the said defendant, has received or made or which have arisen or accrued to it from the aforesaid infringement, together with the damages which plaintiff has sustained by reason thereof;

7. That this cause be referred to a Master hereafter to be appointed by the Court to ascertain and take and report to the Court an account of said profits, gains and advantages and to assess said damages; and pending the appointment of said Master and pending the determination of the appeal herein, if taken, the defendant shall file with the Court on or before the 20th day of each month a report under oath of an officer having knowledge of the facts reciting in reasonable detail the output of product made by defendant by the process herein held to infringe during the preceding calendar month, and the amount of such product sold by defendant during the said preceding calendar month. The first said report shall be filed on or before November 20, 1940 and shall recite such output and such sales for the calendar months of July, August, September, and October, 1940.

8. That a perpetual injunction issue out of and under the seal of this Court directed to the said defendant, U. S. Industrial Chemicals, Inc., restraining the said defendant, its officers, directors, associates, attorneys, clerks, servants, agents, employees and privies, and each of them, from directly or indirectly using or causing to be used, any process for the production of ethylene oxide

embodying the invention claimed in claims 8 and 9 of said Reissue Letters Patent No. 20,370 and from in any way infringing upon either of said claims of said reissue letters patent.

9. That plaintiff, Carbide and Carbon Chemicals Corporation, recover of defendant, U. S. Industrial Chemicals, Inc., its costs to be taxed.

WILLIAM C. COLEMAN,
United States District Judge.

Dated, Baltimore, Maryland, May 2, 1940.

OPINION OF THE UNITED STATES DISTRICT COURT

FOR THE DISTRICT OF MARYLAND

APPEARANCES:

Messrs. WATSON, BRISTOL, JOHNSON & LEAVENWORTH (by L. A. WATSON, Esq., C. W. FAIRBANK, Esq., and CLAIR V. JOHNSON, Esq.), of New York, and Messrs. HERSHEY, DONALDSON, WILLIAMS & STANLEY (by RAYMOND S. WILLIAMS, Esq., and KENNETH EKIN, Esq.), of Baltimore, for the plaintiff.

Messrs. PENNIE, DAVIS, MARVIN & EDMONDS (by DEAN S. EDMONDS, Esq., FRANK E. BARROWS, Esq., and FREDERICK L. BISSINGER, Esq.), of New York, and Messrs. VENABLE, BAETJER & HOWARD (by HARRY H. BAETJER, Esq.), of Baltimore, for the defendant.

COLEMAN, *District Judge*:

This is a patent suit involving a process for the production of ethylene oxide, C_2H_4O, by direct chemical combination between ethylene, C_2H_4,—a hydro-carbon

gas,—and oxygen. Ethylene oxide has various uses, notably when hydrolyzed with water, forming ethylene glycol which is one of the most effective anti-freeze preparations for use in automobiles, and which, therefore, has great commercial value. Soluble in water like alcohol, it is more effective and economical because it does not boil off, like alcohol.

The suit is based upon Reissue Patent No. 20,370, dated May 18th, 1937, which the plaintiff, the Carbide & Carbon Chemicals Corporation, acquired by assignment from one Theodore Emile Lefort, of Paris, France, to whom the original United States Patent No. 1,998,878, was issued on April 23rd, 1935, upon application dated March 22, 1932. This original patent was based upon a process developed by Lefort in France, and there patented March 27, 1931.

The original United States patent, No. 1,998,878, embraced seven claims, and the reissued patent here in suit repeats these seven claims in substantially the same form, but also embraces two additional claims, Nos. 8 and 9, which two are the only ones here in suit, and are virtually identical except in two unimportant respects which will be hereinafter referred to. The specifications in the two patents are quite different in their form,—that is, as to the language employed. Whether any departure in the specifications of the reissued patent from those of the original patent is a material variance, will be considered later.

In both claims 8 and 9, the process set forth for the production of ethylene oxide embraces six steps. We quote claim 8 in full, and have inserted numerals indicating the language in the claim which covers these steps: "The process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene, which comprises (1) forming a mixture containing ethylene and molecular oxygen and (2) conducting said mixture through a confined reaction zone which is maintained at an elevated temperature; (3) controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400°C.; (4) subjecting said mixture in said zone at said

elevated controlled temperature to intimate contact with an active surface catalyst of a material having the catalytic characteristics of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water, and which does not favor the existence in said products of substantial amounts of aldehydes; (5) correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide; and (6) thereafter recovering ethylene oxide-containing product in liquid form."

Shorn of technical phraseology and summarized very briefly, the process may be described as consisting of the passage of a mixture of ethylene gas and air through a tube containing a silver catalyst heated to a control temperature between approximately 150° and 400°. The direct combination of the air with the ethylene produces ethylene oxide. A side reaction also occurs in that there is destructive oxidation of some of the ethylene, producing carbon dioxide and water. It is claimed in the introductory statements of the patent that "This process is charaterized by an entirely unique reaction in which the direct addition of a single oxygen atom to the ethylene molecule occurs without the formation of significant amounts of other oxgenated compounds such as aldehydes." That is to say, the product is claimed to be a notably clean one,—pure ethylene oxide, in readily recoverable form.

The defenses asserted by the defendant, the United States Industrial Chemicals Inc., are both invalidity of the plaintiff's patent and non-infringement. Defendant urges seven grounds in support of its claim of invalidity, as follows: (1) That claims 8 and 9 are vague and functional, and are expressed in terms of result instead of procedure whereby the result is attained; (2) that the specifications of the original Lefort patent were deficient, in that they did not describe the alleged invention in "full, clear, concise and exact terms" as required by Section 4888 of the Revised Statutes (35 U. S. C. A. Sec.

33); (3) that the patent in suit lacks novelty, in that its specifications describe a method known prior to the effective date of the original French application in 1931; (4) that there has been no disclaimer filed by the plaintiff of claims Nos. 2, 4, 5 and 7, wherein there are claimed methods which are not described in the specifications, and also methods, which are stated to be specifically disclaimed in the sworn statement filed by the plaintiff, in furtherance of its application for the reissued patent in suit; (5) that the specifications of the reissued patent in suit describe, and claims 8 and 9 thereof claim an alleged invention different from that described and claimed in the original patent No. 1,998,878; (6) that the original patent No. 1,998,878 was not inoperative for the protection of the alleged invention therein described, and that, therefore, there was no statutory basis for the reissue of the patent; and lastly (7) that there was no inadvertence, accident or mistake in the application for or ground of the original patent No. 1,998,878, and that, therefore, there was no statutory basis for its reissue.

The defendant rests its defense of non-infringement upon two grounds: (1) the usual ground that the process followed by the defendant is materially different from the process described as the invention of the patentee in the original patent No. 1,998,878, and also in claims 8 and 9 of the reissued patent in suit, when properly interpreted in relation to the original patent; and (2) estoppel, which in turn is based upon two grounds: (a) that claims 8 and 9 of the patent in suit can not be given an interpretation sufficiently broad to include defendant's process, because during the prosecution of the application for the original patent No. 1,998,878, in the Patent Office, broad claims were presented but rejected, and the applicant acquiesced in this rejection, cancelled the claims and in connection therewith, stated that he had made an invention of narrower scope involving use of a specific catalyst which was defined in the claims he retained and which the defendant has not used; also that in the application for the reissued patent in suit other broad claims were presented but were rejected; and that the present plaintiff, then owner of this application, acquiesced in the rejection and cancellation of such claims; and (b) that defendant has acquired intervening rights entitling

it to continue to use its process, by reason of the fact that prior to the filing of the application for the reissued patent in suit and also prior to the granting thereof, the defendant had spent large sums of money in research, investigation, development and commercial use of its present process.

While, as above set forth, there are seven separate grounds alleged by the defendant in support of its charge that the reissued patent in suit is invalid, we will start with the third ground, since the conclusion which we reach, as we are now about to explain, with respect to this and the remaining four grounds, especially the fifth and sixth, namely, that they are untenable. renders untenable the first and second grounds also.

With respect to the third ground, namely, that there is lack of novelty in that the specifications describe a method known prior to the effective date of the original French application in 1931, defendant relies primarily, if not indeed exclusively, upon the contention that Lefort is anticipated by two patents to K. P. McElroy: No. 1,253,617 issued January 15, 1918, upon application filed August 2, 1912, and No. 1,308,797 issued July 8, 1919, upon application filed July 26, 1917. Suffice it to say that the overwhelming weight of the credible evidence on this point is to the effect that these patents did not disclose the Lefort process and, in fact, that the processes purported to be disclosed by these patents have never been demonstrated to be operative or of any commercial value. While Professor Taylor, testifying for defendant, stated that in his opinion the McElroy patents did anticipate Lefort; and while the point has been stressed that these patents were not cited to the Patent Office in connection with the Lefort application, we are not convinced by Dr. Taylor's statements. In fact, it is in evidence that in a treatise published by Dr. Taylor in 1926, the statement appears that the reaction claimed by McElroy was impossible,—a contradiction of his testimony in the present case. The McElroy specifications are far broader than the patent in suit; indeed, are devoid of definition of the requisite temperatures, the silver content of the catalyst, etc. These patents, along with others of McElroy, were purchased by the plaintiff company in 1922

for a nominal sum and, as already stated, there is no proof that they have ever been of any practical or commercial value. No witness in the present suit has ever personally tested the sufficiency of the teachings of these patents in direct relation to the patent in suit.

The fourth ground urged by defendant is a failure to disclaim, that is to say, that there was no disclaimer filed by plaintiff of claims 2 and 4, 5 and 7, wherein there are claimed methods which are not described in the specifications, and also methods which are stated to be specifically disclaimed in the sworn statement filed by the plaintiff, in furtherance of its application for the reissued patent in suit.

It is to be noted that this fourth ground relates to claims which are not at issue in the present suit, and which are of a much narrower scope. Suffice it to say that there is no merit in this point, since a patent will not be invalidated for mere failure on the part of the patentee to disclaim prior to some judicial decision upon the subject matter, i.e., upon one or more of the claims, of the patent. After that, the patentee must act with diligence in accordance with the result of the adjudication. Ensten v. Simon, Ascher & Co., 282 U. S. 445, 455; Maytag Co. v. Hurley Co., 307 U. S. 243.

With respect to the fifth ground, namely, that there is a variance between the invention described in the specifications and claims of the original patent and in the specifications and claims 8 and 9 of the reissued patent in suit, the two main points upon which this argument rests are (1) whether the addition of water in the reaction zone is mandatory in either or both of the patents; and (2) whether the specifications in the respective patents are materially at variance as to the type of catalyst to be used.

Our conclusion is that there is no material variance on either of these points. As to water, the contention of the defendant, as set forth in the testimony of its various experts, is to the effect that the addition of water is mandatory under the requirements of the first patent, but not under the reissued patent here in suit. This argument is a highly technical one and is devoid of any

support except such as is to be found in a highly refined interpretation of the antecedent French patents obtained by Lefort, and of certain language employed in the specifications of the United States patents; and also by isolating, erroneously as we think, this language rather than treating it in a practical manner in the light of the language of the respective specifications and claims read and applied as a whole.

Defendant emphasizes the fact that water is not mentioned in the first or introductory paragraph of the reissued patent in its statement of what the process for the production of ethylene oxide consists, whereas, it is referred to in the corresponding paragraph of the original patent; that is to say, the process is there stated to consist mainly of "subjecting ethylene to the simultaneous action of the oxygen, or air and of water, in presence of a catalyzer, and, if need be, of hydrogen." However, with respect to water, the specifications of the original patent are not essentially different from those in the reissued patent.

We find in the original patent that "Water can be admitted in the reaction vessel, either in the liquid state, or as steam. * * * The efficiency of the reaction is increased by diminishing the $C O_2$ which is formed, by introduction, in this reaction, of a suitable quantity of water. * * * From experiments effected by the applicant, it results that, if water is introduced in suitable quantity, the reaction is not only facilitated as above stated, but, in addition, the reaction giving $C O_2$ * * * is checked, owing, as is probable, to the partial pressure of water. * * * The experiments effected by the applicant have shown that, in presence of the catalyzers indicated, water, in the form of steam or not, considerably promotes the reaction insuring the production of ethylene oxide." Then we find in every one of the seven claims of the original patent specific recognition of the existence of water or steam in the reaction zone.

Turning to the reissued patent, the language is not materially different. For example, it is stated in the specifications that the oxidation of ethylene "can be facilitated by suitable dilution of the reaction gases, such as that accomplished by the use of air as the source of

oxygen, and some water or carbon dioxides in addition to that formed can be admitted to the mixture in the reaction zone if desired. Hydrogen may be similarly added. * * * In any case, the ethylene and oxygen are thus reacted simultaneously at the temperatures set forth in the presence of a surface catalyst and of water. * * *." In the added claims in the reissued patent, that is to say, claims 8 and 9, here in suit, the desired reaction is described as taking place "in the presence of water."

On this point it is the weight of the credible evidence as to what actually has been and can be done under the patents that, in the last analysis, should control, and Professor Arthur B. Lamb, testifying on behalf of the plaintiff, has satisfied the Court, through his very clear exposition of the character and results of his laboratory tests, that one skilled in the art, following the teachings of the original patent, will be successful in producing what that patent calls for, and in obtaining such result whether the addition of water be considered as mandatory or as optional, and that he will obtain the same result if he follows the teachings of reissued patent in suit. In other words, what Professor Lamb accomplished by following the original patent proves not only the adequacy of its disclosure as to how to carry out the process successfully, but, in addition, conclusively demonstrates that there is no variance between the subject matter of the original and the reissued patent. It is particularly significant that Professor Lamb never saw the reissued or the third French patent (hereinafter referred to) until after he had testified in the present case. He stated that it never occurred to him that the specifications of the original patent required the addition of water, but that it was advisable to make the addition, in view of the aggregate that was said to facilitate the reaction and, therefore, he did not dry the air. In other words, he decided to use the air either as it came, or with the slight additional amount of moisture that it might pick up from going through the wash bottle.

Summarizing his conclusions, Professor Lamb stated that although he had not read the reissued patent, it necessarily followed from the results of his experiments under the original patent that, even if he had studied the

reissued patent and even if it be assumed that water was specifically omitted as a requirement therefrom, this would not, from the technical chemical point of view, be a material departure.

Defendant has injected into the case a very large amount of testimony with respect to French patents issued to the assignee of Lefort, namely, the Societe Anonyme; Societe Francaise de Catalyse Generalisee, of Paris, as well as certain British patents issued to the same patentee, in an attempt to prove that the process originally conceived by Lefort, in 1931, and patented in France and Great Britain, required the use of water added in more or less large amounts, and that it was not until after the French patentee had applied for a third patent in France in 1933, from which, as it is alleged, the mandatory requirement of his other patents that water be introduced into the reaction chamber was omitted, thereby reducing production costs and explosion hazards, that the plaintiff applied for the reissued patent in suit. In other words, defendant claims that the original United States patent has been reissued as a patent for an invention which had not been made at the time the application for the original was filed; that the reissue is a patent for a different process described in the third French patent and the third British patent, and although based upon a 1931 filing date, is a patent for a 1933 invention.

In support of this contention defendant has gone to great length to analyze the language of the first two French patents, No. 729,952 and No. 739,562, with their various additions, the first of which is stated to be a patent for a process for producing ethylene glycol, and the second for a process for producing ethylene oxide; and also the two corresponding British patents, Nos. 402,438 and 402,749. Reliance is also had upon the assertions of the patentee in the third French patent, No. 771,650, and the third British patent, No. 431,966, to the effect that the invention protected by those patents was, in fact, a new discovery constituting an improvement over the prior patents and embracing certain definite advantages.

But the reissued patent in suit can only be invalidated by evidence far more convincing then what the defend-

ant has produced on this score. Indeed, a critical analysis of the various French patents with their additions, and of the various British patents, is not necessary since they can conclude nothing for our purposes, in view of the testimony already analyzed and which stands, as we think, totally without any successful refutation, given on behalf of the plaintiff by Professor Lamb, and confirmed by Professors Frazer and Stevenson, to the effect that by virtue of their own personal tests made in the light of their knowledge of this branch of chemistry, there is no material difference in the teachings of the original United States patent and the reissued patent in suit with respect to the need for the addition of water in the reaction zone; and that it is only through placing an emphasis, unwarranted by both specifications and claims, upon the reference to water as an ingredient of the process, that any distinction is to be found between the requirements of the respective patents. Indeed, while it is very difficult to reconcile a good deal of the language employed in the various French and British patents,—some of the experts who testified being themselves unable to explain away certain apparent contradictions in the various representations made with respect to the successive claims,—we may assume that under French or British law it may have been appropriate to accord, as was done, the dignity of a patent to the various alleged distinctions and still we are not required to give any conclusive effect to the same in this proceeding.

Finally, whatever weight might otherwise be attached to these foreign patents and the various representations made in connection therewith, is negatived not merely by the contrary findings based upon practical as opposed to merely theoretical considerations, as above explained, but by the fact that the conclusion that the defendant would have us draw from this line of argument, namely, that the application for the reissued patent in suit was inspired by and had no other basis than the third French and British patents, is completely refuted by the unimpeached testimony of plaintiff's patent attorney to the effect that, in his search of the Patent Office records, he found no evidence that applications had ever been made there for patents based upon the third French or British patents, and also, that he had no knowledge of

these patents when he had charge of the application for the reissued patent in its progress through the Patent Office. This supports Professor Lamb's conclusion,—which necessarily follows from the rest of his testimony,—that the third French and British patents represented no actual improvement in the process, by dispensing with the introduction of water vapor into the reaction zone, and thus minimizing, or obviating entirely, explosion hazards by avoiding the necessity of vaporizing large amounts of water.

The Court requested that tests be made in advance of the trial in the presence of representatives of both sides,—confirmed by its insistence upon the appointment of Professor Frazer as a neutral inter-partes expert witness, so that precisely such questions as this one, relating to the use of water might be thoroughly thrashed out in advance of the trial. Yet, after eight days of trial, the defendant produced no witness that had made, or had cooperated with Professor Frazer in making, any inter-partes tests following the teachings of either the original or the reissued patent. So it seems to the Court that the testimony, clear and convincing, of Professors Lamb and Stevenson, and the testimony to the same effect by Dr. Curme, Director of Research for the plaintiff company, supported fully as it is by the inter-partes tests made by the neutral expert, Professor Frazer, must control.

So much for the question of the requirement as to water in the respective patents. Turning to the second question, namely, whether there is nevertheless, a variance in the two patents with respect to the catalytic specifications, we have no hesitancy in concluding, from the weight of the credible evidence, that this question likewise must be answered in the negative.

The gist of defendant's contention is that the specifications and claims are too broad and indefinite with respect to what catalyst shall be used. In other words, the defendant would have us believe that what is specified is merely the use of a silver catalyst that will work, with admonition against using one that will not work,—namely, that the specifications and claims are expressed in terms of the desired result, rather than in terms de-

scriptive of the catalyst itself that will produce such result.

We find this argument totally lacking in merit and, again, the best evidence that this is true is to be found in the testimony of Professor Lamb. Without any knowledge whatsoever of the French patents or of the reissued patent in suit, he followed the teachings of the original patent. He testified that he wanted to avoid trying a number of catalysts and he reasoned that the most efficacious one, and the simplest to prepare, would be one consisting solely of silver because silver was so frequently mentioned in the specifications. He, therefore, used granules of silver oxide, being a compound of silver that would decompose at temperatures within the range specified. He used no carrier, lest he might select one that would be active in decomposing ethylene oxide, and he did not want to assume this handicap. In his first experiment, the conversion of ethylene to ethylene oxide was as high as 28.7%, and in his last experiment (some eleven were made in all), the conversion percentage was as high as 37.7%. His average for all the tests was approximately 33% which, as well as the lowest percentage that he obtained, concededly represents good conversion for commercial purposes. Professor Frazer also used a silver oxide catalyst, but sustained it in filtros. He also used no additional water,—only the water-vapor that entered with the gas mixture, and emphasized his belief that the amount of water was not important. His conversion percentages were somewhat less than Professor Lamb's.

In view of what Professor Lamb was able to do according to the teachings of the original patent, it is idle to contend that the same could not be done by following the teachings of the reissued patent because there is no real variance between the two. In the original patent, twelve different catalysts are set forth, whereas, in the reissued patent there is no such enumeration, the specifications merely prescribing the use of "such a surface catalyst as one composed of silver in finely divided form, and which may be disposed within the tube on a supporting material, or in other known ways." However, in the original patent in two out of the three modi operandi

given as illustrations, as well as in each of the seven claims of that patent, silver, or a mixture containing silver in some form, is specifically referred to. So, this ties up precisely with the specifications of the reissued patent to the effect that a catalyst should be used "composed of silver in finely divided form, and which may be disposed within the tube on a supporting material, or in other known ways"; also, with the further statement, in the reissued patent specifications that "The surface catalyst is preferably silver as stated, but other metals such as bismuth and antimony may be used," and with the fact that each of the first seven claims of the reissued patent (whose language is virtually identical, except for slight transpositions of phrases, with that of the seven claims of the original patent) call for a catalyst composed of silver, or a mixture containing silver.

Defendant stresses the case of Russell v. Dodge, 93 U. S. 46. We find nothing, after a careful examination of this case, in conflict with our decision, because in that case there was obviously not merely a clarification but a change from one invention to a different one.

In deciding as we do, we are not unmindful of the fact that Professor Frazer queried whether the clause in claim 8 (broader than the corresponding one in claim 9) calling for "material having the catalytic characteristics of silver" might be too broad,—i.e. might include *all* future discoveries in this catalytic field,—conceivably catalysts having no silver content whatsoever. However, we do not entertain any doubt as to the validity of the claim on this score, because it must be read in the light of the specifications which, in turn, in their broadest aspect, are to be interpreted as embracing besides silver, only such other metals as are actually enumerated or were actually known, at the time of the original application.

There is one other difference to be noted between claims 8 and 9, i.e., that the latter does, but the former does not, expressly provide for the use of "a heat-absorbing diluent." But this difference is not of material significance, since the use of such a diluent is provided for, optionally in the specifications of both patents, and in

both the same diluents are specified, namely, oxygen, hydrogen, or carbon dioxide.

Defendant claims that Professor Lamb's testimony should be given no credence because it is opposed to the oral testimony of representatives of the Dutch Shell Company, which had had an option to acquire the United States rights in the French patents and in the pending United States application, but which it declined to exercise, because its laboratory tests produced unsatisfactory results; and to certain correspondence introduced in connection with the testimony of these same witnesses, out of which certain parts are taken and stressed as indicating not merely an admission on the part of the French company, the owner of the pending United States application, that it was deficient in its disclosure, but that there was a wilful withholding of necessary specifications. Also, we are asked to reject Professor Lamb's testimony on the ground that his experiments were ex parte.

We are not impressed with any part of this position taken by the defendant. In the first place, defendant can ill afford to attack the experiments and results obtained by Dr. Lamb, or by the other experts for the plaintiff, as being ex parte, since they are not only fully corroborated by tests made by Professor Frazer, pursuant to Court order, in the presence of defendant's representatives, but since defendant has wholly failed to present to the Court *any* experiments whatsoever, conducted by any of its own witnesses pursuant to the requirements imposed in connection with the appointment of Dr. Frazer.

As for the testimony in connection with the Dutch Shell Company's efforts, suffice it to say that we find nothing improper in the correspondence of the French Company above referred to, when considered as a whole, in the light of the business transactions that were involved. Indeed, this correspondence, when so considered, confirms the bona fides of plaintiff's contention that there was real need for clarification of the original United States patent, and that this was the sole basis for the application for the reissued patent and additional claims, and for the changes in the specifications in that applica-

tion over the original application, and for the variations in the additional claims over the claims of the original patent.

Finally, the testimony of the Dutch Shell representatives loses weight because of the admission of one of that company's officials that he was not a disinterested witness in the present litigation, in that, if the patent in suit should be held invalid, his company contemplated entering into the manufacture of ethylene oxide.

Turning to the sixth ground, which is that the original patent was not inoperative for the protection of the alleged invention and that, therefore, there was no statutory basis for the reissue of the patent, this ground is so intimately related to the seventh ground that the two might well be considered as one.

We do not understand that it is necessary, in order to warrant a reissue of a patent, that the original patent be inoperative in the literal sense. The power to reissue a patent may be exercised when the original patent is inoperative in the sense that its specifications are defective or insufficient, or when the claims are narrower than the actual invention of the patentee, provided the error has arisen through inadvertence or mistake, and the patentee is guilty of no fraud or deception; and provided, of course, that the reissue shall be for the same invention as the original patent, as disclosed by the specifications and claims of the original, and that due diligence is exercised in discovering the mistake in the original patent. 35 U. S. C. A. Sec. 64. A delay of two years is too long, in the the absence of very special circumstances. Wollensak v. Reiher, 115 U. S. 96; Topliff v. Topliff, 145 U. S. 156; Webster Co. v. Splitdorf Co., 264 U. S. 463, 466. We find that the plaintiff has met these requirements. It is to be noted that the application for reissue was filed within a year and a half after the original patent was issued.

On the seventh ground, namely, that there was no inadvertence, accident or mistake in the application for or grant of the original patent and that, therefore, there was no statutory basis for its reissue, we conclude that the affidavit by the French attorney,—who was not him-

self a chemist and who sets forth why, acting merely as attorney in Paris and transmitting the papers to the attorney in Washington, he unduly restricted the original application,—adequately disposes of this seventh ground, and we do not consider it is necessary to discuss further the contents of this affidavit. Topliff v. Topliff, supra. The bona fides of the application is further confirmed by the testimony of the attorney for the plaintiff company to the effect that in 1936, after the company's chemists had accomplished satisfactory results in their experiments with the Lefort process, and the company had begun to negotiate for the purchase of the original patent, he examined further into its claims in the light of the original French patents, but without any knowledge of the third French patent, and concluded that the original United States patent was basic; that its claims were too narrow and that, thereupon, he began the preparation of the application for the reissue.

Coming now to the question of infringement, and taking up the first ground of defendant's defense of non-infringement, namely, that the process followed by it is materially different from the process described as the invention of the patentee in the original patent, and also in claims 8 and 9 of the reissued patent when properly interpreted in relation to the original patent, we find no merit in this contention. Plaintiff has graphically set forth in printed exhibits (Plaintiff's Exhibits Nos. 8 and 9) in parallel lines what is required, step by step, under the two claims in issue of the reissued patent, and what is actually admitted as done under defendant's process. Because of the unquestioned similarity, step by step, of the two processes, and because of the almost complete identity of claims 8 and 9, it seems sufficient if we incorporate in this opinion by reference, without copying, the aforementioned comparative analyses of these claims and defendant's process. Again, the Court is unfortunately without the benefit of the results that would have been disclosed had tests been made, as it was understood they would be, on behalf of the defendant, in the presence of plaintiff's representatives, of the process in accordance with the reissued patent and with the process commercially followed by the defendant in its own plant. However, were the results of such tests available, it is

obvious that they would not fail to be corroborative of the conclusion here reached that the processes are virtually identical, and that, therefore, defendant is guilty of infringement. Indeed, in the course of the trial defendant's counsel has admitted that if the presence of water in the reaction zone is found to be optional in either claim 8 or 9, then defendant infringes.

We come then, finally, to the second ground of the defense of non-infringement which, as we have seen, is twofold: (a) that claims 8 and 9 of the reissued patent can not be given an interpretation sufficiently broad to include defendant's process because, during the prosecution of the application for the original patent, broad claims were presented but rejected and the applicant acquiesced in this rejection, cancelled the claims and in connection therewith, stated that he had made an invention of narrower scope, involving the use of a specific catalyst which was defined in the claims he retained; also that in the application for the reissued patent, other broad claims were presented but were rejected; and that the plaintiff, then owner of this application, acquiesced in the rejection and cancellation of such claims; and (b) that defendant has acquired intervening rights entitling it to continue to use its process, by reason of the fact that, prior to the filing of the application for the reissued patent in suit and to the granting thereof, the defendant had, by research, investigation and development, devised and used its present process, involving large expense.

Taking these grounds up in the order stated, the first one may be dismissed forthwith, because it is the same as the fourth contention raised by defendant in its attack upon the validity of the reissued patent, which we have already fully discussed and found to be without merit.

As to the second ground, namely, that of intervening rights, it is difficult to understand how this contention can be seriously made. The principle invoked is well established, namely, that a patentee having consciously accepted with satisfaction a patent of given scope, may not remain idly by and allow his competitor to build up a business outside the scope of the patentee's claims and

thereafter, with a desire to capitalize and take from his competitor what the latter has innocently created or developed, secure a reissuance of the patent so as to embrace the domain of his competitor. See Sontag Stores Co. v. Nut Co., 310 U. S. 281. But it appears that defendant never began to construct its present costly plant until after the reissued patent had been granted; and never began to operate it commercially for the production of ethylene oxide until more than two years later. It is a correct inference from the testimony that prior to the application for the reissued patent, defendant's work on this process amounted to relatively little, but that thereafter, defendant zealously progressed its development of plant and process, with a view to capitalizing what plaintiff had done. It has been testified on behalf of the defendant that more than $1,000,000.00 has been invested in its plant in Baltimore. But this was done by the company's officials with their eyes open to possible results. If they were too hasty or ill-advised with respect to possible conflict between their rights or interests and those of the plaintiff, this is not something that a court of equity can relieve against. Indeed, it is precisely the sort of thing which a court of equity should condemn, not condone.

Defendant's counsel has attempted to make something of the point that the original Lefort patent was not used for five years, and that it is, therefore, to be treated as, in effect, a paper patent. But it issued in 1935, there was competitive bidding for it until plaintiff acquired it in 1936, and the transition of the invention from laboratory demonstration to commercial installation was very rapid. Before the close of the following year plaintiff had erected and was operating a large plant at South Charleston, West Virginia, which now has a yearly output of more than ten million pounds. The French company had licensed the process in England, Belgium, Italy and Russia, and it had also been sought by German interests. In short, the value of the patented process had been recognized commercially in the important European industrial countries.

Defendant stressed the fact that up to the time the application for the reissued patent was filed, namely,

September 25, 1936, its research work had been steadily progressing for some two years, at a cost of over $100,000.00, and that this work was continued during the period that the application was pending, resulting in additional cost to the defendant of more than $50,000.00; also, that during this period, defendant had acquired its United States patent No. 2,125,333 for a catalyst, and had proceeded to develop a plant in which this invention was to be practiced. Defendant relies upon Sontag Stores Co. v. Nut Co., supra, and Otis Elevator Co. v. Atlantic Elevator Co., 47 F. (2d) 545, a decision of the Circuit Court of Appeals, 2nd Cir. Defendant insists that its operations should be considered as entitled to protection up to the date of the application for the reissue, even if not to the date of its granting, and even though all such operations were entirely in the experimental and development stage, as distinguished from commercial operations. The cases relied upon do not so decide, nor do any to which we have been referred. In the Sontag case, the facts are quite different from those here presented. There, the Court assumed, without deciding, the correctness of the lower Court's conclusion that the enlarged reissue claims were valid and infringed. Also, the reissue was applied for within two years after the granting of the original patent. But here the analogy to the facts in the present case ceases, because in the Sontag case, the infringer actually began to use the accused machine within a year after the original patent was granted, and his manufacturer had begun to make and sell like machines two or three months thereafter; whereas in the present case, as already pointed out, the defendant not only did not enter upon the commercial production of ethylene oxide until more than two years after plaintiff had obtained its reissued patent, but never started the construction of its plant until after the reissue had been granted. With respect to the Otis Elevator case, the reissue there was not applied for until more than seven and one-half years after the original patent had been obtained, three of which were excused on account of the intervention of war, but the additional delay was not. The intermediate advance in the particular art there involved, i. e., that relating to stopping devices for electric elevators, was also an important

factor. So, this case, likewise, is entirely different upon its facts from the case at bar.

For the aforegoing reasons a decree will be signed holding claims 8 and 9 of the patent in suit both valid and infringed.

WILLIAM C. COLEMAN.

FINDINGS OF FACT AND CONCLUSIONS OF LAW

Findings of Fact

1. The patent in suit involves a process for the production of ethylene oxide C_2H_4O, by direct chemical combination between ethylene, C_2H_4,—a hydro-carbon gas,— and oxygen. Ethylene oxide has various uses, notably when hydrolyzed with water, forming ethylene glycol, which is one of the most effective anti-freeze preparations for use in automobiles, and which, therefore, has great commercial value. Soluble in water like alcohol, it is more effective and economical because it does not boil off, like alcohol.

2. The suit is based upon Reissue Patent No. 20,370, dated May 18th, 1937, which the plaintiff, the Carbide & Carbon Chemicals Corporation, acquired by assignment from one Theodore Emile Lefort, of Paris, France, to whom the original United States patent No. 1,998,878, was issued on April 23rd, 1935, upon application dated March 22nd, 1932. This original patent was based upon a process developed by Lefort in France, and there patented March 27th, 1931.

The original United States patent, No. 1,998,878, embraced seven claims, and the reissued patent here in suit repeats these seven claims in substantially the same form, but also embraces two additional claims, Nos. 8 and 9, which two are the only ones here in suit, and are virtually identical except in two unimportant respects which will be hereinafter referred to. In both claims 8

and 9, the process set forth for the production of ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene, embraces six steps: (1) forming a mixture containing ethylene and molecular oxygen and (2) conducting said mixture through a confined reaction zone which is maintained at an elevated temperature; (3) controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400° C.; (4) subjecting said mixture in said zone at said elevated controlled temperature to intimate contact with an active surface catalyst of a material having the catalytic characteristics of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water, and which does not favor the existence in said products of substantial amounts of aldehydes; (5) correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide; and (6) thereafter recovering ethylene oxide-containing product in liquid form.

The process may be briefly described as consisting of the passage of a mixture of ethylene gas and air through a tube containing a silver catalyst heated to a control temperature between approximately 150° and 400°. The direct combination of the air with the ethylene produces ethylene oxide. A side reaction also occurs in that there is destructive oxidation of some of the ethylene, producing carbon dioxide and water.

3. The reissued patent in suit is valid.

4. Claims 8 and 9 of the plaintiff's patent are not so vague and functional as to render said patent invalid, but describe the procedure and not merely the result attained thereby.

5. The specifications of the original Lefort patent were sufficient in that they described the alleged invention in "full, clear, concise and exact terms" as required by Section 4888 of the revised Statutes (35 U. S. C. A. Sec. 33).

6. There is no lack of novelty in the patent in suit, for the specifications describe a method unknown prior to the effective date of the original French application in 1931. The two patents to K. P. McElroy (No. 1,253,617 issued January 15, 1918, upon application filed August 2, 1912, and No. 1,308,797 issued July 8, 1919, upon application filed July 26, 1917) did not disclose the Lefort process and, in fact, the processes purported to be disclosed by these patents have never been demonstrated to be operative or of any commercial value. The McElroy specifications are far broader than the patent in suit; indeed, are devoid of definition of the requisite temperatures, the silver content of the catalyst, etc. These patents, along with others of McElroy, were purchased by the plaintiff company in 1922 for a nominal sum.

7. There is no variance between the invention described in the specifications and claims of the original patent and in the specifications and claims 8 and 9 of the reissued patent in suit.

8. With respect to water, the specifications of the original patent are not essentially different from those in the reissued patent. Professor Arthur B. Lamb, testifying on behalf of the plaintiff, has satisfied the Court, through his very clear exposition of the character and results of his laboratory tests, that one skilled in the art, following the teachings of the original patent, will be successful in producing what that patent calls for, and in obtaining such result whether the addition of water be considered as mandatory or as optional, and that he will obtain the same result if he follows the teachings of the reissued patent in suit. In other words, what Professor Lamb accomplished by following the original patent proves not only the adequacy of its disclosure as to how to carry out the process successfully, but, in addition, conclusively demonstrates that there is no variance between the subject matter of the original and the reissued patent. This testimony is confirmed by the testimony of Professor Frazer, the inter-partes neutral witness, and also by that of Professor Stevenson and Dr. Curme, witnesses for the plaintiff.

9. The application for the reissued patent in suit was not inspired by and was not based on the third French

and British patents. Plaintiff's patent attorney, in his search of the Patent Office records, found no evidence that applications had ever been made there for patents based upon the third French or British patent. It is also clear that he had no knowledge of these patents when he had charge of the application for the reissued patent in its progress through the Patent Office.

10. There is no variance in the two patents with respect to the catalytic specifications. In the original patent, twelve different catalysts are set forth, whereas, in the reissued patent there is no such enumeration, the specifications merely prescribing the use of "such a surface catalyst as one composed of silver in finely divided form, and which may be disposed within the tube on a supporting material, or in other known ways." However, in the original patent in two out of the three modi operandi given as illustrations, as well as in each of the seven claims of that patent, silver, or a mixture containing silver in some form, is specifically referred to. So, this ties up precisely with the specifications of the reissued patent to the effect that a catalyst should be used "composed of silver in finely divided form, and which may be disposed within the tube on a supporting material, or in other known ways"; also, with the further statement in the reissued patent specifications that "The surface catalyst is preferably silver as stated, but other metals such as bismuth and antimony may be used," and with the fact that each of the first seven claims of the reissued patent (whose language is virtually identical, except for slight transpositions of phrases, with that of the seven claims of the original patent) call for a catalyst composed of silver, or a mixture containing silver.

11. The clause in claim 8 (broader than the corresponding one in claim 9) calling for "material having the catalytic characteristics of silver" must be read in the light of the specifications which in turn in their broadest aspect are to be interpreted as embracing besides silver only such other metals as are actually enumerated or were actually known at the time of the original application.

12. Although claim 9 does and claim 8 does not, expressly provide for the use of a "heat-absorbing diluent,"

this difference is not of material significance, since the use of such a diluent is provided for optionally in the specifications of both patents, and in both the same diluents are specified, namely oxygen, hydrogen, or carbon dioxide.

13. The correspondence of the French company, then owner of the French patents and the pending United States application, with the Dutch Shell Company, which had an option to acquire the United States rights in the French patents and in the pending United States application, discloses that there was real need for clarification of the original United States patent. This need afforded a basis for the application for the reissued patent and additional claims, and for the changes in the specifications in that application over the original application, and for the variations in the additional claims over the claims of the original patent.

14. The original patent was sufficiently inoperative for the protection of the alleged invention to warrant the reissue of the patent. While it may not have been inoperative in the literal sense, the original patent was inoperative in the sense that its specifications were defective or insufficient and the claims were narrower than the actual invention of the patentee. Such error arose through inadvertence or mistake, the patentee being guilty of no fraud or deception. The error was discovered with due diligence, and the application for the reissue was filed within a year and a half after the original patent was issued.

15. Defendant infringes plaintiff's reissued patent in suit.

16. The process followed by defendant is not materially different from the process described as the invention of the patentee in the original patent and in claims 8 and 9 of the reissued patent, as becomes obvious from an examination of Plaintiff's Exhibits Nos. 8 and 9 setting forth comparative analyses of the two processes step by step, which analyses are incorporated by reference in these findings of fact.

17. Defendant acquired no such intervening rights as would estop plaintiff from asserting defendant's in-

fringement. Defendant never began to construct its present costly plant until after the reissued patent had been granted; and never began to operate it commercially for the production of ethylene oxide until more than two years later. Prior to the application for the reissued patent, defendant's work on this process amounted to relatively little, but thereafter, defendant zealously progressed its development of plant and process, with a view to capitalizing what plaintiff had done. Defendant's investment of more than $1,000,000 in its plant in Baltimore was made by the company's officials with their eyes open to possible conflict between their rights or interests and those of the plaintiff.

18. The original Lefort patent is not to be treated as, in effect, merely a paper patent. It issued in 1935, there was competitive bidding for it until plaintiff acquired it in 1936, and the transition of the invention from laboratory demonstration to commerical installation was very rapid. Before the close of the following year plaintiff had erected and was operating a large plant at South Charleston, West Virginia, which now has a yearly output of more than ten million pounds. The French company had licensed the process in England, Belgium, Italy and Russia, and it had also been sought by German interests. In short, the value of the patented process had been recognized commercially in the important European industrial countries.

Conclusions of Law

1. Plaintiff has full title to the reissue patent in suit.

2. The Court has jurisdiction in the suit.

3. Claims 8 and 9 of plaintiff's reissue patent No. 20,370 are valid.

4. The specifications of the original Lefort patent do not fail to describe the alleged invention in "full, clear, concise and exact terms" as required by Section 4888 of the Revised Statutes (35 U. S. C. A. Sec. 33).

5. Plaintiff's patent is not invalid for failure of plaintiff to file disclaimer of claims 2 and 4, 5 and 7, which

are not at issue in the present suit and which are of a much narrower scope; for a patent will not be invalidated for mere failure on the part of the patentee to disclaim prior to some judicial decision upon the subject matter, i. e., upon one or more of the claims, of the patent.

6. While, under French or British law, it may have been appropriate to accord, as was done, the dignity of a patent to the various alleged distinctions in the specifications of the French patents and their additions, and of the British patents, still this Court is not required to give any conclusive effect to the same in this proceeding.

7. The clause in claim 8 (broader than the corresponding one in claim 9) calling for "material having the catalytic characteristics of silver" does not render claim 8 invalid on the ground that this clause is too broad in that it might include all future discoveries in this catalytic field,—conceivably catalysts having no silver content whatever,—because this clause must be read in the light of the specifications which in turn in their broadest aspects are to be interpreted as embracing besides silver only such other metals as are actually enumerated or were actually known at the time of the original application.

8. The original patent was sufficiently inoperative for the protection of the alleged invention to afford a statutory basis for the reissue of the patent. It is not necessary, in order to warrant a reissue of a patent, that the original patent be inoperative in the literal sense.

9. The power to reissue a patent may be exercised when the original patent is inoperative in the sense that its specifications are defective or insufficient, or when the claims are narrower than the actual invention of the patentee, provided the error has arisen through inadvertence or mistake, and the patentee is guilty of no fraud or deception; and provided, of course, that the reissue shall be for the same invention as the original patent, as disclosed by the specifications and claims of the original, and that due diligence is exercised in discovering the mistake in the original patent. A delay of a year and a half after the original patent was issued is not too long.

10. Defendant infringes claims 8 and 9 of plaintiff's reissue patent No. 20,370, in that the process followed by defendant in producing ethylene oxide is not materially different from the process of claims 8 and 9 of plaintiff's reissue patent No. 20,370.

11. A patentee having consciously accepted with satisfaction a patent of given scope, may not remain idly by and allow his competitor to build up a business outside the scope of the patentee's claims and thereafter, with a desire to capitalize and take from his competitor what the latter has innocently created or developed, secure a reissuance of the patent so as to embrace the domain of his competitor.

12. If in making large investments the officials of the defendant company were too hasty or ill-advised with respect to possible conflict between their rights or interests and those of the plaintiff, this is not something that a court of equity can relieve against. Indeed, it is precisely the sort of thing which a court of equity should condemn, not condone.

13. Defendant's operations cannot be considered as entitled to protection up to the date of the application for the reissue or to the date of its granting, since all such operations were entirely in the experimental and development stage as distinguished from commercial operations.

WILLIAM C. COLEMAN,
United States District Judge.

October 30, 1940.

UNITED STATES DISTRICT COURT

DISTRICT OF MARYLAND

CARBIDE AND CARBON CHEMICALS CORPORATION, *Plaintiff*, *vs.* U. S. INDUSTRIAL CHEMICALS, INC., *Defendant.*	CIVIL ACTION FILE No. 249

Order

It is hereby ORDERED by the District Court of the United States for the District of Maryland, this 20th day of May, 1940, that Dr. J. C. W. Frazer of Baltimore, Maryland, is hereby appointed to serve as a neutral expert witness in this cause to advise the Court with respect to reissued letters patent in suit No. 20370, the process of the Defendant in its relation thereto and the disclosures of the prior art in their relation to the said reissued patent in suit.

IT IS FURTHER ORDERED that the said Dr. Frazer shall in advance of the trial of this cause make such tests and experiments in respect to any and all of the foregoing matters as he may deem desirable and appropriate, affording, so far as is feasible and as he may deem desirable, to representatives of both parties and their counsel the right to be present at any such tests and experiments; and shall, at the request of either party attend such inter-partes tests and demonstrations respecting any of the foregoing matters as may be made prior to the trial of this cause; and may suggest to either party that that party make such tests and demonstrations as he may consider pertinent to any of the issues of the case and at which tests or demonstrations he and representatives of the other side shall have the right to be

present; so that he shall be in a position to advise the Court with respect to all of the aforesaid and his conclusions thereon at the trial, which advice shall be given in the form of sworn testimony in open court and subject to cross examination by each of the parties, and to the objections of counsel.

AND IT IS FURTHER ORDERED that the fees and expenses of the said Dr. Frazer shall be fixed by the Court and paid in the first instance equally by the parties, and finally as part of the taxable costs herein.

AND IT IS FURTHER ORDERED that the parties shall promptly furnish to Dr. Frazer the following documents:

1. The reissue letters patent No. 20370 in suit.

2. The original patent No. 1998878.

3. Defendant's statement as to its process, a copy of which has been furnished to Plaintiff.

4. A corresponding statement of Plaintiff's operations, a copy of which is to be supplied to Defendant at the same time.

5. Copies of the prior art set up as anticipations in Defendant's answer as amended on which Defendant will rely at the trial, together with translations of such as are not in the English language, it being understood that nothing herein shall abridge the right of either party to refer to other publications at the trial as indicating the state of the prior art.

6. File wrapper of reissue patent 20370.

7. File wrapper of original patent 1998878.

8. French patents Nos. 729952 and 739562 and the Additions thereto, together with translations of them.

9. British patents Nos. 402438 and 402749.

WILLIAM C. COLEMAN,
United States District Judge.

Dated: May 20, 1940.

AMENDMENTS TO PARAGRAPH 6 OF DEFENDANT'S ANSWER

6. Further answering, defendant avers that said Reissue Letters Patent No. 20,370 were and are invalid and void for each of the following reasons:

* * * * * *

g. Because plaintiff was fully informed and fully aware that said Reissue Letters Patent contains claims, in particular claims 2, 4, 5 and 7 thereof, for subject matter which is not disclosed in the specification thereof, and plaintiff has unreasonably neglected and failed to disclaim said claims or any one or more of them.

(Amendment filed September 29, 1939)

h. Because plaintiff was fully informed and fully aware that said Reissue Letters Patent contain claims, in particular claims 4 and 5 thereof, for subject matter which was specifically disclaimed by the patentee Lefort in an oath signed and sworn to by him under date of March 30, 1937, and filed in the Patent Office in furtherance of his application for the said Reissue Letters Patent, and plaintiff has unreasonably neglected and failed to disclaim said claims or either of them.

(Amendment filed April 19, 1940)

TESTIMONY OF DANIEL PYZEL

Thereupon——

DANIEL PYZEL, a witness of lawful age, produced on behalf of the defendant, having been first duly sworn, was examined and testified as follows:

Direct Examination.

By Mr. Edmonds:

Q. Mr. Pyzel, you have stated your name. And will you please give your place of residence? A. My place of residence is Piedmont, California.

Q. And do you spend much of your time in New York? A. I spend about half of my time in New York, and half of my time in California.

Q. And are you in the employ of an industrial organization? And if so, will you state in what capacity? A. I am in the employ of the Royal Dutch Shell, the Shell Union Oil Company, and the Shell Chemical Company. Do you want the capacity?

Q. Yes, will you state that? A. I am Vice-President and Director of the Shell Union Corporation; I am Vice-President and Director of the Shell Chemical Company; and a director of the Shell Development Company.

Q. (By Mr. Edmonds) What is the relation of the Shell Development Company to the Shell Union Oil Company? A. The Shell Development Company is a company that occupies itself with research work and patents.

Q. That is its field of operations. What is its relation to the Shell Union Oil Company? A. The Shell Development Company does the research work for the operating companies.

Q. Do these companies with which you are connected, and more particularly the Shell Union Oil Company, have any relation with industrial organizations abroad? And if so, will you state what they are? A. The Shell Union—all these Shell companies in this country are subsidiaries of the Royal Dutch Shell Company in Europe. This same combination of Royal Dutch Shell has utilities in Europe. One of them is the Bataafsche Petroleum Maatschappy over there.

Q. May we refer from now on to that company as B. P. M.? Will that be convenient? A. It will be much more convenient.

Q. Had you finished? A. The B. P. M. is the subsidiary of the Royal Dutch Shell, which is in charge of all the production and refining and operation of plants, and is also in charge of the research and development work, as far as it is being done and carried out in Europe.

Q. Now, would you state whether or not B. P. M. in Europe corresponds to the Shell Development Company

in the United States, in the Dutch Shell organization? I do not mean specifically, but in a general way? A. Yes, the development department of the B. P. M. in Europe does the same work as the Shell Development Company in this country, yes.

Q. And is there any more direct connection, as, for instance, an ownership connection, between Shell Development and B. P. M.? A. The Shell Development in this country is owned 50 per cent. by the Shell Union Oil Corporation in New York, and 50 per cent. by the B. P. M. abroad * * * [311-313].

* * * Q. Did your Dutch Shell companies or any one of them at any time have an option to acquire the application for the Lefort patent? The original patent of which the reissue herein suit is a reissue? A. Yes, sir.

Q. At about what time did you get that option and how long did the option continue? A. The option was acquired in, if I remember right, about April, 1933, and ran to August 1, 1933, and then was extended until the 31st of December of that year.

Q. How did the matter first come to your attention? A. It came first to our attention through correspondence with our friends in The Hague.

Q. Now, will you proceed in narrative form, if you will, and state what happened from that time on, giving a narrative of it? A. A research group investigated it. This patent was offered by Societe Francaise de Catalyse Generalisee, and our research group sent some of their men over to the laboratories of this French company to have this process demonstrated to them and to get acquainted. The reports on these investigations were in due course sent to us,—— * * * [316-317].

* * * It was then reported by our associates in Holland that we could obtain a license agreement under those patents under certain conditions * * * [318].

* * * And at the same time they sent us a full report on the experimental work that was shown by the French inventors to the chemists of our laboratory.

The conclusion of these reports was that the value of the invention was completely in the catalyst employed,

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NOTE: Figures in brackets refer to pages of typewritten record.

and, therefore, before we decided we could go into this proposal on this side we thought it was necessary to investigate whether the patent, which at that time was only an application, would give us sufficient protection if we were going to use that patent. The difficulty was that in these experiments nothing was told about the character of the catalyst, and as our patent department here was of the opinion that unless that catalyst was properly disclosed in the patent it would not give us sufficient protection, we set our men to work to find out whether, with the disclosure in the patent application and in the French patents, it would be possible to operate this process.

This work was carried on more or less simultaneously at our Holland laboratory and in our laboratories in California, with the result that with data, as they were in the patent and in the French patent and in the American patent application, we found it was impossible to carry out the process.

In order to have sufficient time to investigate that, our friends on the other side asked for an extension of the option from the 31st of August, 1933, to the 31st of December which, after some difficulty, we obtained from the French company.

The French company did not at that time understand, and wrote us so, why the option should be extended. They said, "If you want to investigate we have to admit —we have to warn you that the data given in the patent application"—— * * * [318-320].

* * * As I said, the French company wrote us that they thought it was a waste of time because with the data in our hands in the patent application, we would not be able to get the results that the process could give if we had all the details of how to make or how to use that patent.

The Court: I do not understand that last. Will you state that again?

Mr. Edmonds: Will you restate that to the Court so that the meaning will be unmistakably clear?

The Witness: The French company did not think it necessary in the beginning to give an extension of

the option because the option could only be, in their opinion, an extension could only be for the purpose of allowing us to do more research work, and they said in their letters that further research work would not give us any result as, with the data in our hands, that is, the statement in the French patents—I mean in the American patent application, it would be impossible to obtain the results.

The Court: You mean that the owners admitted you could not get results with their own work, with their own formula?

The Witness: Yes, sir, the disclosures in the patent application.

By Mr. Edmonds:

Q. Did they tell your representatives who went to the French company what the catalyst was? A. No, they refused to tell that.

Q. Your representatives tried to get that information, did they not? A. We did.

Q. And it was after that unsuccessful effort to find out what the catalyst of the French company was and after finding the information in the patent application itself insufficient, that the French company wrote to you what you said they wrote? A. Yes, sir.

Mr. Edmonds: Does that clear that up, your Honor?

The Court: Yes, I think it does. The inference to be drawn from that, I suppose, is that they wanted you to take it or leave it, so to speak? In other words, if you wanted to enter into a license agreement you would take the patent, but they were not going to disclose anything more unless you did enter into an agreement with them. That's what it comes down to, isn't it?

The Witness: Yes, they only wanted to give us the full information after we should have signed an agreement, for which we would pay so much and pay a certain royalty.

The Court: Yes.

Mr. Edmonds: But involved in that is an admission that the description in the application was insufficient.

Q. Now, will you proceed with the narrative? You were speaking of the research work that went on in Holland and in this country. A. We continued our research work and used various kinds of silver catalysts in order to—in an effort to duplicate the work that the French company had shown us, but both in our laboratory in Amsterdam and our laboratories in California we were not successful. Numerous silver catalysts were used but they had practically no effect and with no formation of ethylene oxide, and this was an important point for us because it was the opinion of our patent department, our company's patent department here, that in that case a license under the patent of the French company would not protect us sufficiently, and that was one of the reasons we finally broke off the negotiations.

Q. Now, was the research work at The Hague continued during the fall and winter of 1933? A. It was continued in 1933 and in the beginning of 1934, and apparently the work was continued even after the option officially lapsed and the work was continued in this way: that a certain process was tried out in the laboratory of the French company and on a certain gas mixture containing ethylene that we had available in one of our plants in California, and for that reason the work was continued.

Q. Was there some further contact with the French company along in the winter of 1933 and 1934, I mean, work with them right at their plant? A. Yes. These experiments with the gas of the composition that we had available in California were continued on in their laboratories.

Q. And was a report on that written? A. A report on that was written, a full report.

Q. And did the work at The Hague go on beyond that, in point of time, and continue even after that visit to the French company? A. Yes, our laboratory in Amsterdam continued to work to find a suitable catalyst for this process.

Q. Did the laboratory succeed in doing that finally? A. It succeeded in finding and Van Peski obtained a patent on that special catalyst in this country.

Q. Will you tell us to the extent that you know about it whether or not water was one of the reactants put in the reaction chamber in the method of the Dutch Shell Company to comprise the process as described in the application of Lefort for French and United States Patents? A. Do you mean in the experiments made together with the French company?

Q. Yes. A. Yes, in all those experiments water in quite considerable quantities was used.

Q. For instance, in the experiments made at the French company's plant in the winter of 1933 to 1934 at a time when, as you said, you sent some Shell gas over there for use in the demonstration, was water one of the reactants put into the reaction chamber then? A. Yes, sir, water was added in considerable quantities, if I remember right, eighty or one hundred grams of water per one hundred litres of gas.

Q. Roughly, can you give what percentage of the whole mixture introduced into the reaction chamber is represented by the things you have just given, what percentage of total volume? A. The volume of the steam generated from that water that was added was equal or more than the volume of the other gases, so that the mixture of gases and steam would constitute more than 50% of water vapor.

Q. I want you to identify some documents, if you will. You spoke of the initiation of this matter of an option to your company to get a license under this Lefort U. S. application. Will you state whether that document I hand you bears on that? A. Yes, it does.

Q. Is that a copy of a letter written by you? A. It is a copy of a letter written by me to Mr. Masthoff of the company's office in the Hague. He is in charge of chemical developments * * * [320-327].

* * * Q. The original went to Holland presumably. It is there, if it is anywhere? A. Yes, it is there, it should be there * * * [327-328].

Mr. Edmonds: * * * We will offer in evidence the copy just introduced or identified by the witness as Defendant's Exhibit No. 42.

(Copy of letter written by Mr. Pyzel to Mr. Masthoff marked "Defendant's Exhibit No. 42.")

By Mr. Edmonds:

Q. Mr. Masthoff, as far as you know, is he the chief of research of B. P. M. in Holland? A. Whatever is left of it at present.

Q. Did you get a reply to the letter which you sent and which has just been offered in evidence? A. I guess I got a reply in which Mr. Masthoff tells us what they had done so far with the French company.

Q. Is that the original of the reply? A. Yes, that is.

Q. Addressed to you and signed by Mr. Masthoff? A. It is addressed to me and signed by Mr. Masthoff.

Q. And you received it shortly after the date that appears? A. I received it on July 8th, 1933.

Q. You will notice at the end there reference is made to a number of enclosures that were to be forwarded with it and presumably were, one of them is a copy in French of the U. S. Lefort application and I ask you whether the document I now hand you is that particular enclosure received by you with that letter? A. Yes, it is.

Q. I believe the letter also makes reference to a copy of the U. S. application in English and I will ask you if the document I now hand you is the copy of the application in English? A. Yes, it is.

Mr. Edmonds: I offer these three documents in evidence, the letter of July 28, 1933 from Mr. Mastoff to Mr. Pyzel as Defendant's Exhibit No. 43, the copy in French of the application of Lefort as Defendant's Exhibit No. 44 and the copy of the application in English as Defendant's Exhibit 45.

(The three documents indicated, letter of July 28th, 1933 from Mr. Masthoff to Mr. Pyzel, copy in English of the application for the Lefort patent and copy of the application in English marked "Defendant's Exhibits 43, 44, and 45" respectively.)

Q. Now, this letter which has just been marked Defendant's Exhibit 43 refers also to a report No. 5205 as enclosed with the letter and I ask you whether the document which I now hand you is that report? A. That is the document.

Mr. Edmonds: I will offer this report in evidence as Defendant's Exhibit No. 46.

(Report No. 5205 marked "Defendant's Exhibit No. 46.")

Q. Will you state just what this report forming Exhibit 46 is? A. It is a report by Doctor Langedijk of the company in Amsterdam of his visit to the Societe Francaise de Catalyse Generalisee, to their laboratory where this process of direct oxidation of ethylene to ethylene oxide was demonstrated to him.

Q. At the conclusion of the visit of Doctor Langedijk to the S. F. C. G. Company, was any arrangement made for a further meeting of representatives of the B. P. M. and the French company in Holland? A. Yes, there was.

Q. Did that meeting take place? A. The meeting took place.

Q. Is this document I now hand you a report of occurrences at that meeting? A. Yes, it is.

Q. Is that document, that particular one, the one that was sent to you with this letter of June 28th, Exhibit 43? A. Yes, it was.

Mr. Edmonds: I offer in evidence the report of the meeting of the representatives of the French company and the B. P. M. as Defendant's Exhibit No. 47.

(Report of meeting of representatives of French company and B. P. M. marked "Defendant's Exhibit No. 47") * * * [328-331].

* * * Q. Now, I hand you another document and ask you to state what that is? A. This is a letter from Mr. Masthoff in the Hague to me.

Q. Dated what and received by you when? A. That was received on July 20th, 1933.

Q. Now, that letter states at the end of it, it lists at the end of it certain enclosures that went with it. Did you receive those enclosures that went with the letter? A. Yes. The first enclosure is a letter from the French company to our friends in The Hague; the second enclosure is the reply of our friends in The Hague to the French company and the third enclosure, which is a memorandum of our Patent Department in The Hague on the subject.

Mr. Edmonds: I will offer in evidence this letter of July 6th, 1933 just identified by the witness, together with two of the three enclosures; that is the letter from the French company of June 18th, 1933 and the letter in reply to that dated July 5, 1933, and I would like to state that the third of the enclosures we have here and he is welcome to it at any time——

But being a report of the Patent Department of the B. P. M., it seems to me that the plaintiff here might have a proper basis for objecting to it, so I am not offering it in evidence; but they can see it; and if they have no objection to it it will be offered in evidence.

Mr. Watson: I will take it now, if you please.

Mr. Edmonds: Will you mark this exhibit, the letter of July 6, 1933, as Defendant's Exhibit No. 48.

(Letter dated July 6, 1933, marked "Defendant's Exhibit No. 48.")

Mr. Edmonds: I also offer in evidence translations into English of these two letters in French which accompanied Defendant's Exhibit No. 48.

(Translations of letters marked "Defendant's Exhibit No. 49.") * * * [334-336].

* * * Q. (By Mr. Edmonds) Look at the document that I now hand you, dated July 17th, and state whether or not you received that from the B. P. M., and with it the enclosures that are annexed to it (handing papers to witness). A. Yes, I did.

Mr. Edmonds: I offer that in evidence as Defendant's Exhibit No. 50.

(Letter dated July 17th, and annexed enclosures, marked "Defendant's Exhibit No. 50.")

Mr. Edmonds: The two letters annexed to this Exhibit No. 50 are in French; and so I present translations of them as Defendant's Exhibit 51.

(Translations marked "Defendant's Exhibit No. 51.")

Q. (By Mr. Edmonds) Look at the document I hand you now, July 25th, with one letter attached to it (handing papers to witness). Did you receive that letter at about that time? A. Yes, sir, I did.

Q. And the enclosure annexed to it? A. Yes.

Q. That particular paper was annexed as the enclosure? A. Yes.

Mr. Edmonds: I offer that in evidence as Defendant's Exhibit No. 52.

(Letter dated July 25th, and annexed enclosure, marked "Defendant's Exhibit No. 52.")

Mr. Edmonds: I also offer in evidence a translation of the letter in French, as Defendant's Exhibit No. 53.

(Translation marked "Defendant's Exhibit No. 53.")

Mr. Edmonds: Now, after you received these several letters what did you do next? And did you confer with any one connected with the research department of Shell development?

A. Yes, the research department of the Shell development, to do a certain amount of work to find out whether with data in the French patents—I mean, the American patent application, they could carry out this reaction.

Q. And did Shell development proceed under Mr. Williams' direction to perform that research? A. Yes, they did.

Q. And I suppose that was generally under Mr. Williams' charge, if so, who had charge of it under him? A. Mr. Williams is the head of the laboratories, and he instructed Dr. Groll to undertake this work with the group that he had. Dr. Groll was the head of a group of chemists.

Q. And the work was done and the report of it made to you? A. Yes, it was.

Q. Do you remember in what period the work was done? A. The work was done in August, 1933.

Q. Now, I ask you what is the document I now hand you? A. This is a monthly progress report on the work done in the month of August, 1933, by the group of chemists working under Dr. Groll in our laboratories in California.

Q. And you saw it at about the date it bears? A. Oh, yes * * * [345-349].

* * * Mr. Edmonds: I offer in evidence the title page of this report No. 1759, and page 68 of it, that being the only page of it that relates to this matter of ethylene oxide. But the balance of the report counsel can examine and satisfy themselves that it has no relation to anything else * * * [349-350].

* * * Q. Is that the original document, that mimeographed copy? A. Yes, that's the way in which our laboratory reports are issued because of the large number of copies that are required.

Q. On some of these reports you put right on the title page to whom copies are to go, do you not? A. Yes, sir.

Mr. Edmonds: I would be glad to keep that copy of the full report so that if counsel will agree that photostat copies may be substituted, I would like to do that.

I offer in evidence the two pages of the report, the title page and page 68 of the report itself, as Defendant's Exhibit 54.

(Title page and page No. 68 of Report No. 1759 marked in evidence, "Defendant's Exhibit 54.")

Q. Now, will you state what this document is I hand you? A. This is a copy of a cablegram which I sent to our friends in The Hague.

Q. On the date that it bears, August 29? A. Yes, sir.

Q. And in that cablegram did you record very briefly a preliminary opinion that you had arrived at by that date? A. Yes, I did.

Mr. Edmonds: I offer in evidence a cablegram just referred to by the witness as Defendant's Exhibit 55.

(Cablegram of August 29 marked, "Defendant's Exhibit 55.")

Q. Will you look at this document and tell me what it is? A. This is a copy of a cable I sent to the B. P. M. to do the work about a month later.

Q. Now, is it in that telegram that you referred to sending over some Shell gas for experimentation? A. Yes, sir.

Q. And was that done? A. That was done.

Q. At or about that time did you receive any inquiry from B. P. M. about something further that B. P. M. wanted to know for their use in the experimentation that they were conducting?

(Witness does not answer.)

Mr. Edmonds: Well, if you don't recall just what I refer to I will ask you to look at the document I now hand you, and I will offer in evidence the cablegram of September 28, 1933, which the witness has identified as Defendant's Exhibit 56.

(Cablegram from Mr. Pyzel to B. P. M. dated September 28, 1933, marked, "Defendant's Exhibit 56.")

A. Yes, we asked our laboratory in Amsterdam to do some further work on these catalysts in order to find out any way to make the catalysts so that they would work, and the reply from our friends in The Hague was that they had already started experiments in Amsterdam for the reasons explained, and they asked us to telegraph the composition of the catalysts which we used in our experiments in the laboratories in California.

Mr. Edmonds: I will offer in evidence copy of this telegram of October 2 as Defendant's Exhibit 57.

(Copy of cablegram of October 2 marked, "Defendant's Exhibit 57.")

Q. And what is the document I now hand you? A. This is a copy of a letter which I wrote to B. P. M. in

The Hague in which various silver catalysts with which we had experimented in the laboratories in California are described.

Q. That is your compliance with the cable request that you just referred to? A. Yes.

Mr. Edmonds: I offer in evidence copy of Mr. Pyzel's letter of October 6th as Defendant's Exhibit 58.

(Letter from Mr. Pyzel dated October 6th marked, "Defendant's Exhibit 58.")

Q. Look at the letter which I now hand you dated October 2, 1936, and state what that is. A. This is a letter which Mr. Masthoff of the B. P. M. in The Hague sent to me in California.

Q. Received by you in the regular course of the mail? A. Yes, received ten days after it was mailed in The Hague.

Q. And as received did it have attached to it the enclosure that is attached to it now? A. Yes, it did.

Mr. Edmonds: I offer in evidence this letter of October 2, 1933, together with the letter in French attached to it as an enclosure, as Defendant's Exhibit 59.

(Letter from B. P. M. to Mr. Pyzel of October 2, 1933, together with letter in French marked, "Defendant's Exhibit 59.")

Mr. Edmonds: I offer in evidence a translation of the letter in French, which is the enclosure with this letter of October 2, 1933, this translation to be, Defendant's Exhibit 60.

(Translation of letter in French enclosed with letter of October 2, 1933, marked, "Defendant's Exhibit 60.")

Q. Will you look at the copy which I now hand you and state what that is? A. This is a copy of letter which I sent to the B. P. M. at The Hague on October 19, 1933.

Mr. Edmonds: I offer this copy in evidence as Defendant's Exhibit 61.

(Letter from Mr. Pyzel to the B. P. M. in The Hague on October 19, 1933, marked, "Defendant's Exhibit 61.")

Q. I now hand you a letter dated October 20, 1933. Will you state what that is and also what the enclosure with it is? A. This is a letter sent by B. P. M. in The Hague to me in San Francisco.

Q. Received by you in the regular course of the mail? A. And received by me ten days after it was mailed.

Q. And what is the enclosure? A. The enclosure is a report of a discussion between Mr. Francon of this French company and some of the gentlemen of the B. P. M. in The Hague.

Mr. Edmonds: I offer in evidence this letter of October 20, 1933. that the witness has just identified, together with the copy of their report in answer to it as an enclosure, as Defendant's Exhibit 62.

(Letter of October 20, 1933, from B. P. M. to Mr. Pyzel, together with enclosure, marked, "Defendant's Exhibit 62.")

Mr. Edmonds: The enclosure with that letter is in French, so I will offer the translation of it as Exhibit No. 63.

(Translation of letter in French enclosed with letter of October 20, 1933, marked, "Defendant's Exhibit 63.")

Q. Now, at or about the time that we have now come to, did Mr. Masthoff come to this country? A. Yes, he came to this country.

Q. And did you meet with him and confer on various subjects, including this one? A. Yes, I did.

Q. Did you two collaborate in writing anything in the way of instructions to the B. P. M.? A. Yes, we did.

Q. Look at the copy of the letter of November 3, 1933, which I now hand you and state what it is. A. This is a copy of a letter sent by Mr. Masthoff from The Hague who at that time was in California, to Mr. Jacobson of the B. P. M. in The Hague, on the subject of ethylene oxide.

Q. In that letter, towards the end, does that recall to your mind any reference as to how you and Mr. Masthoff conferred on this matter? A. We discussed whether this process for which the French company offered us a li-

cense would be of any use for our purposes especially in comparison to other processes of making Ethylene Glycol and our conclusion was that the other processes were preferred.

Q. I take it that the original of that letter was signed by Mr. Masthoff? A. This letter was signed by Mr. Masthoff, yes, but it was the result of a discussion between Mr. Masthoff and me and he refers to that in the first paragraph of this letter and also in the second from last paragraph.

Q. The letter represented your views at the time as well as his? A. Yes, it was our joint opinion.

Mr. Edmonds: I offer this copy in evidence as Defendant's Exhibit No. 64.

(Letter referred to marked "Defendant's Exhibit 64.")

Q. Now, will you look at the document I now hand you and state what that is? * * * [351-360].

* * * The Witness: This is the condensed report on the work of the research laboratory in Emeryville, covering the last nine months of 1933.

By Mr. Edmonds:

Q. This particular volume is a rather large one covering a great many subjects other than ethylene oxide? A. Yes, it does, it covers all the work that has been done during those nine months in the laboratories.

Q. I wish you would state whether or not it is a fact that the only references to this ethylene oxide matter contained in the report are on pages 42, 44, 45 and 46. Is that a fact? A. Yes, it is.

Mr. Edmonds: I will offer in evidence photostat copies of the title page of this report and of pages 42, 44, 45 and 46 * * * [360].

* * * Q. Now, will you look at the binder which I now hand you and state what is contained in that? A. This is a binder of the laboratory in California containing monthly reports of the work done in the laboratory in

Amsterdam on the subject of production of ethylene oxide.

Q. What period do the monthly reports cover? A. October, it starts in October of 1933, December, 1933 and goes on to January, 1935.

Q. Did you receive copies of these reports as they were issued? A. Yes, sir.

Q. You personally? A. I got these reports personally and distributed them from my office.

Q. So they went to Emeryville in addition to going to you? A. Yes.

Mr. Edmonds: I offer in evidence this collection of reports just identified by the witness as Defendant's Exhibit 66.

(Collection of reports of work done in laboratories in Amsterdam marked "Defendant's Exhibit 66.")

By Mr. Edmonds:

Q. Can you state from the papers which I now hand you when these monthly reports were received by you? A. These monthly reports, for instance the monthly reports of the Amsterdam Laboratory of October and November 1933 were received here in California in March, 1934. For the work done in January and February of 1934 we received them in June of 1934. And for the work done in March and April 1934 we received the reports in September, 1934.

Q. Now, will you identify this document which I hand you which has on the front of it the original of a letter dated May 28th, of 1934? A. This is a document which was received in California on June 12th, 1934 and contains the reports of the work done in the presence of two of the chemists in the Amsterdam Laboratories, Mr. Al and Mr. Langedijk in the laboratories of the French company in order to find out whether the certain gas mixture which is produced at one of our plants in California and which contains etheylene may be converted into or could be used for the purpose of producing ethylene oxide by this process. These experiments were

made in January, 1934, from January 9th to January 24th and this is a full report on the work that was done at that time.

Q. Did you explain what gas was used in making these tests covered by the report? A. It was what we call an ethylene fraction, which is produced in the Shell Chemical and Ammonia plant in California and which contains around 50% of ethylene.

Q. Was it that particular gas referred to in some of the cablegrams that you identified as being sent over? A. Yes, it was.

Q. And a copy of this report came to you at about the time it was submitted, did it not? A. Yes, this work was done on my request.

Q. And did you examine the report? A. I did.

Q. And you have retained until today your familiarity with it? A. Yes, sir.

Q. And on that basis what can you say about the water that was introduced into the reaction chamber in the course of the operations covered by the report? A. All this work was being carried on in the laboratories of the French company. The procedure followed was as indicated by the inventor and in all the runs that were made and all the efforts that were made to obtain good results, to improve results, always a considerable quantity of water was used.

Q. Can you express that in figures, not exactly, but reasonably accurately, as, for instance, the percentage of the volume of water which went into the reaction chamber? A. Well, it is in the direction of 80 or 100 grams and sometimes more and sometimes a little less per 100 litres of gas, and that works out always to at least 50% of the mixture of gases that goes into the reaction chamber as water vapor.

Q. This experimental work at the French plant covered by this particular report was some nine months later than the work recorded in one of the other reports that you identified here this morning? A. Yes, the first report was on work done if I am not mistaken, April 1933 and this is January, 1934.

Q. Is there any statement in this report which you have in front of you about the amount of water that went into the reaction in the course of this 1934 experiment as compared with the amount that went in in the 1933 experiments of nine months earlier. A. The above statements in this report on that point——

Q. Just state the general purport of them? A. Here, for instance, per 100 litres of this mixture of Shell point gas, Shell point is the name of the ammonia plant they have here, about 100 grams of water had to be used. There are indications in this record that the reduction of the reaction temperature has a favorable effect.

Q. Isn't there some statement there that the amount of water introduced in 1934 was a little less? A. I remember that there is a statement that the amount of water introduced during these experiments was less than what was used in the experiment of 1933, but where it is exactly, I don't know.

Mr. Edmonds: I offer in evidence a photostat copy of this report which the witness has just identified as Defendant's Exhibit 67 and will ask counsel to agree that this photostat copy may be accepted in lieu of the original, it being understood that the original will be here for examination at any time.

(Photostat copy of report above mentioned marked Defendant's Exhibit 67.)

Mr. Watson: You don't have to keep repeating that, we will always take copies.

By Mr. Edmonds:

Q. Now, I think you spoke of the Van Peski patent. Is this the Van Peski patent, 2040782, which you referred to? A. Yes, this is the patent.

Q. That covers the work done at the B. P. M. Laboratory along about 1934? A. Yes, it does.

Mr. Edmonds: I offer in evidence a copy of the Van Peski patent 2040782.

(Copy of Van Peski patent 2040782 marked "Defendant's Exhibit 68") * * * [362-368].

* * * Q. Now, you mentioned Dr. Groll, Mr. Pyzel, as one of the research men in the laboratory. When did he leave Dutch Shell and where did he go? A. I do not quite exactly remember when he left.

Q. Well, say a year ago? A. Say about two years ago, and he went to Germany.

Q. And he has been wholly disconnected from the Shell Company since then? A. No, he first worked in Germany with an oil company that belonged to our company.

Q. Oh, I see, but so far as you know he is in Europe now, and probably in Germany? A. Oh, yes.

Q. And you had something to do with the effort to get Dr. Langedijk to come over here for this litigation, did you not? A. Yes.

Q. About a month ago? A. Yes.

Mr. Edmonds: Now, I want to go into this other matter concerned with the objection that was presented by counsel. Where are those four reports?

Q. I wish you would state in some detail what, if any, system was adopted for the interchange of technical information between the Shell Development Company and B. P. M., pointing out to what extent the system that was adopted included the preparation of reports on items of consequence.

The Court: Now, that's the question I ruled out, isn't it, about an hour ago?

Mr. Edmonds: Yes, sir, because its bearing at the time did not appear, but its bearing does now, because the plaintiff has objected to the receipt in evidence of these documents, and they are receivable, as I see it, only if I meet the conditions of the statute, and this is designed to present the proof required by the statute making those documents admissible.

The Court: Well, gentlemen, as I understand it, the witness has testified as a high official of the Shell Company, that his company asked these technicians to look into the French patent and give the results of what they thought of it, whether it was worthy of being licensed,

and you have put in that correspondence, and the witness has stated that in effect the reports say, "We do not think so much of the patent." In other words, they recommend against their doing anything about licensing it.

Now, that's the sum and substance of the testimony. Now, I do not see any need of going into the internal relations of this or that company, or its relations with some other company. I do not see that that helps at all. I am letting in, subject to the renewal of the objection, I am letting in the prima facie proof and the correspondence, and it has been summarized by the witness. I understand you want somebody else to talk in more detail about the report. It seems to me that that is ample. I am not interested, for many reasons, in relation to this case, and I see no need to go any further into the history of the way these companies work; I am not interested in how they work, I am interested in how this thing works, the particular process.

Mr. Edmonds: May I ask two questions, using the words of the statute, by way of offering to produce that information which I conceive to be necessary to make these documents admissible? All I want to do is to comply with the conditions of the statute. The statute says if it shall appear that it was made in the regular course of any business, and that it was the regular course of such business to make such memorandum or record at the time——

The Court: But Mr. Pyzel has already explained in some detail that he ordered them, the experiments, or that they were ordered by his general direction, and that they were carried out, and he has discussed in a brief way what they amounted to. I do not see that you need anything else. I do not want to go in detail, it wouldn't throw any light on the facts, and that's what we want to get to, and we want to get there as briefly as possible.

Mr. Edmonds: I am faced, may it please the Court, with the fact that counsel for the plaintiff has put on the record an objection to the admissibility of these documents.

The Court: Well, I overruled the objection with the right to renew it later on, but I am now admitting everything that you put in, I don't know whether it has any real probative value or not, but I am letting it in on the basis it may have.

Mr. Edmonds: May I just make this statement for the record, then: that the defendant offers to prove by this witness that all the documents identified by him were made in the regular course of the business?

The Court: He has already said that. I recall that you asked him such a question—maybe not in those exact words—and Mr. Pyzel has stated that, and stated it several times, that's why I am letting it in.

Mr. Edmonds: I thought that was covered by the question which was ruled out initially, and which I repeated.

The Court: I am letting it on the basis that Mr. Pyzel had direction or he was one of the high officials who had the general direction over these laboratory experiments which were ordered either directly or indirectly by him for the purpose of determining whether his company would take out a license, would take out a license under these French patents.

Now, you have introduced a very elaborate chain of documents showing, as I understand it, that they went fully into this thing and made elaborate experiments and concluded that it was not worth taking out a license and therefore the matter was turned down by the technicians of the Dutch company, which Mr. Pyzel represents. Now, he has stated all that, and I am letting it in because of his position in the company and his general direction over these laboratory experiments, and that's all you need.

Mr. Watson: May I offer one more comment? For instance, here is one of these exhibits. We are getting a tremendous volume of them. So far as they go in I trust they will go in as physical exhibits that do not have to be reproduced in 50 copies on an appeal record.

Mr. Edmonds: Oh, I guess we can agree about that later on.

The Court: In other words, if, for whatever reason, Mr. Pyzel is not now requested to elaborate any further on these reports, I should say that your direct examination was about concluded.

Mr. Edmonds: It is, so far as I am concerned, in all respects except this one thing that gives me some concern.

Your Honor has admitted these documents, but suppose we were in a court of appeals and I make my argument based on these documents, and counsel then says, "Those should not have been admitted at all; my objection there was a proper one because there was no compliance with the conditions of Section 8695."

The Court: I think there has been a substantial compliance.

Mr. Edmonds: Well, could we abbreviate the matter by allowing me to make a statement on the record that we offer to prove by this witness that all of these documents were made in the regular course of business of his companies, and that it was the regular course of that business to make all of these memoranda and records?

The Court: Yes, you can ask him that question. I thought you had done that.

Mr. Watson: My objection goes beyond that point. My objection persists, I do not care what the language is of the statute, I think it is impossible to believe that this Court can take such documents from a witness who neither wrote them nor received them, nor understands what they were at the time they were done, and have them evidence of the facts recited in them. My objection is fundamental.

Mr. Edmonds: You can abbreviate this a great deal by saying you do not contend that these documents were not made in the regular course of business, or that it was not the regular course of business to make such records.

Mr. Watson: I can not admit that, Mr. Edmonds; some of this work was done in Holland, in Europe;

what is the materiality? What it has got to do with this case I can not imagine.

Mr. Edmonds: Well, that is another matter.

The Court: Well, I will allow the witness to answer the question, and I understand his answer is, Yes. It has already been put to him.

The Witness: That's right.

By Mr. Edmonds:

Q. You have stated that all of these records that you have identified here today were made in the regular course of the business of your Shell Development Company and the B. P. M.? A. Yes, sir.

Q. And was it the regular course of business of those companies to make just such memoranda and records? A. Yes, it was.

Q. And was it throughout the years prior to 1933, and has it been ever since? A. Yes, sir.

Mr. Edmonds: That's all.

The Court: Now, do I understand you are following this witness with some witness who did do some of the experimenting?

Mr. Edmonds: Yes, sir, all experiments done in August, 1933, and covered by the report which Mr. Pyzel identified, the report of the one for 1933; that man is here and we propose to put him on the stand.

The Court: Well, for the present, then, subject to your right to renew the objection as to the ruling, I think it would be more appropriate to limit the admission of the documents to the part that the witness present who is going to testify to them knows about. I think the letters are admissible, but as far as the reports go, I think they should be limited, in the production of them, to what the following witnesses or witness, who will follow, will speak about, and that partially meets Mr. Watson's objection.

Mr. Edmonds: I think the witness only knows about the work that is covered by one of the records——

The Court: Well, I think it is a more satisfactory way to proceed, and then you can renew your objection as to the other report following the hearing of the testimony with respect to the witness who made the particular laboratory experiment on the basis of the one he knows about. That is very much narrower than Mr. Watson means.

I think there is great weight to what Mr. Watson said. You cannot use a strict technical—you could not and should not require someone to rest without having an opportunity to ask the man who made the experiment why he used this or that quantity or this or that thing. It is not like a financial report where two and two make four. I think the statute has that sort of thing more in mind perhaps.

Mr. Edmonds: It certainly was directed among other things to entries but it goes way beyond that.

The Court: Yes, but it has to be given a reasonable interpretation.

Mr. Edmonds: And the Courts have already carried it way beyond that as, for instance, the special Court that ruled on the Manton case, where it received in evidence records of checks.

The Court: I will have to limit the admission as just stated. I think one of the grounds for being perhaps a little more liberal than usual is because of present war conditions and inability, the admitted inability to introduce in cases over here involving foreign work witnesses that would ordinarily be called but cannot for the time being on account of the conditions on the other side, but for the time being I am going to limit the admissibility to the correspondence and the one report about which you say you have a witness who can testify of his own knowledge * * * [381-393].

* * * Mr. Edmonds: What am I to understand from that, that the report on the test made with Shell gas in France in January of 1934, that is excluded? That is the report this witness said he studied and was familiar with. The detail in that report is like the detail that goes into an account book in a bank. No one person can be produced

to identify any of these details because they were made by——

The Court: Someone signed that report or rendered it, didn't he?

Mr. Edmonds: Probably a number of people collaborated in writing it.

The Court: The records of the company would show who submitted it, wouldn't they?

Mr. Edmonds: They would show various people who did the work, contributed their part to making up the report.

The Court: My present ruling is that I am excluding all of the scientific reports for the time being which cannot be followed up by testimony of some persons, someone or more persons who actually performed the experiments referred to or made some part of the final report bearing there on.

By Mr. Edmonds:

Q. Mr. Pyzel, in these monthly reports which you identified of the work at the B. P. M. laboratory from October, 1933 to April, 1934, will you tell us whether or not Doctor Langedijk did some of the work covered by these reports? A. Yes, I am quite certain that he did.

Mr. Watson: Are you guessing? Did you see him do it?

The Witness: No, but he reports that.

The Court: Is he here?

The Witness: He is the man we cannot get here on account of the war conditions.

By. Mr. Edmonds:

Q. He is the man you expected or thought you had arranged to get here, but he could not come? A. No.

Q. I see some of these reports indicate that some of the work was performed by a Mr. Al? A. Yes.

Q. Is he available for use here? A. No.

Q. Do you know where he is? A. In Holland.

Q. Some of the reports indicate that part of the work was done by Mr. Smithuysen? A. Smithysen.

Q. Where is he, do you know? A. In Holland.

Q. Who is Doctor Visser, whose name appears on some of the reports? A. He is one of the chemists in the laboratory in Amsterdam.

Q. Did all three of these men continue in the laboratory up to a month ago as far as you know? A. Yes, sir.

Q. How do these reports compare with reports which you have received from time to time since 1933 and reports which you received years prior to that?

Mr. Watson: I object to a question of that kind. It is calling for a conclusion of the witness. If the thing was material, Mr. Edmonds would have to produce these other records.

The Court: I sustain the objection and, Mr. Edmonds, I think I have made my ruling clear and will have to adhere to it. You can have an exception and you can renew your objection later on, but I will limit the introduction for the time being to the correspondence that has general relation, I understand, to this whole subject, and which has a bearing even though it may be indirect at times upon this one report or such other reports if there is more than one, which is going to be referred to by one or more witnesses who naturally had something to do with making it up.

By Mr. Edmonds:

Q. I want to ask you, you said the option to take a license under the U. S. Pending Application of Lefort ran up to December 31, 1933. Did your companies allow that option to expire? A. Yes, it had expired.

Q. And further work was done in January covered by one of these reports. Was that done under an extension of the option? A. Yes, I cannot tell you exactly but the option was informally extended because the work

was done together with the French people in their laboratories.

Q. And extended without any writing on the subject at all? A. None at all.

Mr. Edmonds: That is all.

Mr. Watson: Mr. Bissinger, you are familiar with these exhibits. Can you give a list of those which are admitted under the Court's ruling?

Mr. Edmonds: All are admitted except two, this one here and the big one here * * * [394-399].

The Court: What are the numbers of the ones that are excluded?

Mr. Edmonds: The numbers of the ones excluded are 66 and 67. Those two, we understand, are excluded. What is covered by this one in general form, this monthly progress report, is work which is also covered by the detailed report not yet presented on which this other witness will testify.

The Court: Let's get on, gentlemen.

CROSS EXAMINATION.

By Mr. Watson:

Q. Where were you born, Doctor Pyzel? A. Excuse me, I am not a doctor.

Q. Where were you born, however? A. In Holland.

Q. How long have you been in this country? A. Twenty-two years.

Q. Are you here under subpoena? A. No, I am not.

Q. You are not here under any kind of Court order? A. No, I am not.

Q. You came of your own free will? A. Yes.

Q. Who are your higher officers in this country? A. Well, I would say Mr. Vanderwoude, the president of Shell.

Q. Did he tell you to come here? A. No * * * [399-400].

* * * Q. Do you know the Shell Company is interested in the outcome of this suit? A. We are not as far as I——

Q. Answer my question. A. Do I know?

Q. Yes. A. Not directly.

Q. What do you mean by not directly? You would like to go into the business of making ethylene oxide? A. We would like to be able to do that, yes.

Q. And you would like to do it by a catalytic process of passing ethylene and air over silver? A. We would like to do it by a catalytic process of direct oxidation.

Q. Using silver? A. I don't care, anything that is a good catalyst.

Q. If the Lefort patent is held invalid, it is your intention to go into that business? A. We may.

Q. How long have you known of the existence of the Lefort U. S. Patent? A. The Lefort U. S. Patent?

Q. Yes. A. I think about from the time it issued.

Q. How long about the Lefort Reissue patent, the one here in suit? A. Well, I can't exactly tell. I get all these patents in the form of abstracts over my desk and if they especially interest me I look at them and, if not, I only look at the abstract as they issue.

Q. When did your option to acquire the American rights expire? A. Officially on the 31st of December, 1933.

Q. Do you know your companies tried to open negotiations to get a license under the Reissue patent? A. I don't remember.

Q. Are you a chemical engineer skilled in the process of catalysis? A. I am a chemical engineer. I know something of the process of catalysis. How skilled I am, I don't know.

Q. Would you call yourself a skilled worker in the field of catalytic processes in 1933 for example? A. I think I know more about it than a lot of other chemists.

Q. That is not my question. We will proceed faster if you will answer the questions. What had been your experience up to that time in processes of gas phase reactions by catalysis? A. One of our experiences is a large ammonia plant in California which is used as a catalytic process.

Q. As a Haber process? A. No, and where purification of gases takes place by catalytic processes.

Q. What are they? A. The removal of carbon dioxide from the mixture of hydrogen and nitrogen and admixtures of ammonia from admixtures of hydrogen and nitrogen.

Q. Is that the extent of your experience? A. That is the extent of a very intimate experience. There are further processes, oil, in which an enormous amount of work on catalysis has been done and on which I have always been kept informed and which I had to study in the course of my duties.

Q. That is prior to 1933? A. Yes, I started in 1929.

Q. You were informed by the Societe Francaise de Catalyse Generalisee that the Lefort process included the use of a silver catalyst, weren't you? A. Yes, sir.

Q. And you were informed that the process was conducted at a temperature of 150 to 400 degrees centigrade, weren't you? A. Yes.

Q. And you were informed also that the French companies patent rights in foreign countries had been acquired in England, for example, weren't you? A. I don't remember.

Q. When did you find out that the Distillers Company Ltd. of England owned the British patents to the Lefort process? A. Now, I remember, we received information from the company here in New York.

Q. So you knew that the British Distillers Company was operating the Lefort process in England? A. Well, whether they were actually operating I don't know, whether that was in the information.

Q. And the German I. G. acquired the corresponding patent rights in Germany to the Lefort process, do you know that? A. I didn't know that * * * [401-405].

* * * Q. Do you know whether the I. G. Company in Germany bought the Lefort patent rights for Germany? A. I don't know.

Q. Were you ever informed that that was a fact? A. I don't quite remember. We were informed of such things but they were often incorrect as information.

Q. Were you informed that the I. G. Company was negotiating to acquire the German rights? A. I don't quite remember that with certainty.

Q. Would you remember that if it did appear from some of the records which you had just referred to? A. Oh, yes, if they are in the records.

Q. But you don't remember without looking at the records, is that what you mean? Now, referring to the experiments conducted by the Shell Development Company, I presume that is an experimental company? A. Yes, in this country.

Q. And that work was done in August of 1933? A. Yes, sir.

Q. The only work done by the Shell Development Company in this country to test the Lefort process was in that month of August, 1933? A. Yes.

Q. And that is the one that is summarized in this report marked Exhibit—what is the number, is it here? Is it the one I hand you?

Mr. Edmonds: Identify it for the moment as report No. 1759.

Mr. Edmonds: I think that no copy of the—what is your Exhibit 54?

The Clerk: That was a monthly progress report of work done in August, 1933.

Mr. Edmonds: Yes, the marking of the copy of Exhibit 54 was deferred because no photostatic copy was available at the time. That copy is now available, and

is being marked "Defendant's Exhibit 54," with the agreement that that copy may be used in lieu of the original.

(Monthly progress report on work done in August, 1933, by chemists working with Dr. Groll, marked "Defendant's Exhibit No. 54.")

Q. (By Mr. Watson) Mr. Pyzel, was the decision of the American Shell Company not to exercise its option to buy the Lefort patent rights in this country, so far as that decision was based upon experimental test work conducted in this country, based upon that experimental test work in August, 1933? A. Partly.

Q. What other experimental work in this country? A. No other experimental work in this country.

Q. So that, answering my question, so far as it was based upon work done in this country, that work was exclusively work done in Emeryville, in August, 1933? A. Yes, sir.

Q. And that work was reported to you by Dr. Groll? A. Dr. Groll, Dr. Williams, in the laboratory.

Q. Well, I show you Sheet 2 of Exhibit 54, which is stated to be the only one on the subject. A. But you asked for the report. The report includes that.

Q. So that the only written report was that signed by Dr. Groll? A. Yes, sir.

Q. Now, on your familiarity with catalytic processes, did you yourself direct or supervise this work done at Emeryville in August, '33? A. No, I did not.

Q. Did you see it done? A. I don't remember.

Q. Do you know what it consisted of? A. I read the reports.

Q. Well, answer me, what did the work consist of? A. I tried to find out whether——

Q. That is the object of the work. I mean what was the procedure, what materials were used? A. Ethylene, air, oxygen, steam, various kinds of catalysts.

Q. Well, now, was silver used? A. Certainly.

Q. Silver by itself? A. Certainly, silver by itself was used.

Q. How was that silver made? A. In different ways.

Q. How was it prepared as a catalyst? A. It was prepared by various methods.

Q. Well, name them. A. Well, I will have to go back to the records, because I could not remember that far back.

Q. I am asking you, do you know? A. No, I don't remember everything that happened seven years ago in the laboratory.

Q. Well, did you know it then? A. It was a very large laboratory; and it is impossible for any man to remember everything in detail, what happened.

Q. Did you yourself ever try to make a silver catalyst? A. No.

Q. To conduct this process? A. No, sir.

Q. If you were proceeding to do so, as of that date, in the first place, would you take silver? A. If I know that process that I want to check is using a silver catalyst, of course, I would start with silver.

Q. All right. If you know that the process you are wanting to check says that you will use silver on a support or not, you would try it on a support and not on a support, would you not? A. That I don't know. I probably would.

Q. If you tried it on a support or carrier, you would pick out an inert carrier wouldn't you? A. Not necessarily.

Q. Why not? A. Because it is very difficult to say whether a support is inert or not.

Q. Isn't it a simple matter to test it? A. Not as simple as you think it is.

Q. Well, can't you pass ethylene and air mixture over it? A. Oh, yes.

Q. At the temperatures involved? A. I can take one support and another support, both supposed to be inert, and get completely different results.

Q. Do you mean to say that it is difficult to find out whether a support material is inert when ethylene or air, or ethylene oxide is passed over it, at a temperature of between 150 or 400 degrees? A. Yes, it is difficult to determine those things.

Q. You mean so difficult that you have got to try it? A. Well, even so; even if you try it, it is still difficult. It is a most difficult subject that we are talking about.

Q. If you are going to test silver as a catalyst, would you not try to use a support so that when you got your silver on it you would know you were testing the silver and not the support? A. That would be the object of the support, because the support might make a big difference on the behavior of the silver as a catalyst.

Q. Assuming you are testing a process where you are told the silver is a catalyst, not the support is the catalyst, but the silver is the catalyst, then, would you not pick out a support that was inert so that you would know when you put the silver on it you were testing the silver and not the support? A. It is not that simple. Silver can be in all kinds of forms, and the character of the support on which the silver is precipitated or carried or whatever it is, can have an enormous influence on the silver as the catalyst.

Q. I understand you to say it is difficult. Do you mean to suggest that it would be nonsense to try to use an inert support when you are attempting to test the behavior of silver as a catalyst? A. Oh, certainly not. I did not say that. The only thing I said, it is very difficult to know whether the support is really inert or not.

Q. Yes, but it would be obviously desirable to use an inert support, since you are going to test the silver? A. We are going to test the silver on a support or not on a support, and we tried to find out what is the result of that. It is very difficult. If you talk about silver as a catalyst that helps very little; it depends on exactly the shape in which that catalyst is, on what it does, and you have to regulate the shape, and we don't know too much about it. It is exactly how that catalyst is made that is of importance, and that's what we try to find

out. If you say silver is the catalyst you have said nothing, but if you want to have certain results you have to define exactly how to make it because that silver in another form may give you completely different results.

Q. Is that a peculiarity of the Lefort process or is that true of all catalytical processes? A. It is very dangerous in all catalytical processes to make conclusions before you try them.

Q. So that you mean to indicate there is nothing peculiar about this Lefort process, you are making a general comment applicable to all catalytical processes? A. Yes, sir.

Q. So that you think any patent on a catalytic process is totally insufficient in disclosure unless it tells you how to prepare a catalyst in detail? A. It has to give you at least some indications of how to prepare it.

Q. The use of the term "silver" is not enough, in your opinion? A. No.

Q. How about "silver in finely divided form"? A. No.

Q. When you are told to use silver, the skill in the art would be sufficient, would it not, to attempt to use this silver in an active surface form? A. It may be active for one process and inactive with another process, or it may be inactive in the wrong direction. A silver catalyst has been used for the oxidation of ethylene oxide to produce formaldehyde. That's not what you want.

Q. You are expressing a conclusion and I am asking for facts. Was it not well known in the art how to prepare an active surface silver catalyst in 1933? A. Not in this general form, no. It all depends on what type process you want. The surface may be active for one process and may be inactive for another process.

Q. Yes. Did you study the literature or do you know whether your laboratory workers studied the literature in 1933 on the preparation of silver catalysts? A. I expect they did.

Q. I am asking you whether you know it? A. No.

Q. It would be a normal, sensible thing to do, wouldn't it, study the literature? A. I should think it would.

Q. And if the literature indicated suitable inert carriers it would be naturally normal to select one, wouldn't it? * * * [407-417].

* * * A. Yes, sir, it would be, I would say, very remarkable to not try silver on an inert carrier, if you know it is inert.

Q. And the carrier should possess certain mechanical characteristics, isn't that true? A. It may be.

Q. Well, it would be obviously desirable, would it not, to select a carrier of some mechanical strength? You wouldn't want one that crumbled? A. No.

Q. You would want it to have some mechanical strength? A. Yes.

Q. And you would want it to have some kind of surface so that the silver would adhere to it? A. Yes.

Q. And it would obviously be desirable to use it in a form so that when you passed gases through it it would not impede the gas flow, isn't that true? A. You mean it should be granulated?

Q. Preferably. You wouldn't want to use dust? A. No.

Q. And you would want it in such form that the gas would not tend to channel through? A. Yes, sir.

Q. Do I understand you to say that silver has been used in the oxidation of ethylene oxide to formaldehyde? A. As far as I know, yes.

Q. Did you ever do that? A. No.

Q. Did you ever see it done? A. No. It is the subject of a patent.

Q. All right, what patent? A. I don't know.

Q. You don't know what date? A. No, I don't remember the numbers and dates of all the patents I have seen.

Q. Well, having turned to the literature, we will assume in 1933, and having followed the obvious common

sense of selecting an inert carrier which has some mechanical strength and was granular and had a surface to which the silver would adhere, do you know whether the literature taught as one suitable method of depositing silver on such a support, the use of silver nitrate precipitated to silver oxide and reduced, say, at 200 degrees Centigrade with some hydrogen? A. Certainly. It is one method of getting silver precipitated on some carrier.

Q. Do you know whether your experimenters in Emeryville in August, 1933, ever followed that procedure? A. I don't know. I don't remember all these figures, all of these experiments.

Q. What was the price of the option the Shell Company got in the United States? A. I think it was a million and a half francs, but I am not quite sure about it. I would have to look that up.

Q. They paid that for the option? A. No—oh, excuse me—I don't think we paid anything for the option. It was the option to take it, to buy or to use it for a million and a half francs, plus the royalty.

Q. Thirty per cent royalty? A. No.

Q. As far as you know it may have been a 30 per cent royalty? A. No, I don't know. I will have to look at those figures * * * [418-421].

* * * The Witness: Here is a letter from our friends in The Hague to the French company in which they say, in the translation, "Two million francs and a royalty of one franc per kilogram, which you ask we feel bound to tell you we are unable to take into consideration any payment of such magnitude."

Q. And what's the date of that? * * * A. 5th of July.

Mr. Watson: That's all right.

The Witness: Here is a letter from our friends in The Hague to the French people.

Q. And that refers to the sale of the American rights? A. Yes, sir.

Mr. Edmonds: You are also referring to this cablegram of August 29, 1933?

The Witness: Yes, this refers to that date. Here I suggest a—

"Further study of process from technical, economical and patent viewpoint before making recommendation for agreement binding us to definite royalty and cash payments. Consider anyhow royalty far too high and cash payment exorbitant, but can only make definite counter proposals after complete study as mentioned above."

Mr. Edmonds: Have you got the translation of the letter of July 5? The witness read from the paragraph at the bottom of page 1, Exhibit 49.

By Mr. Watson:

Q. It appears from Exhibit 49, then, that you rejected the terms offered for the American rights even before you conducted tests, is that correct? Exhibit 49 was the letter written by somebody from The Hague, in July, July 5, 1933 (exhibiting to the witness)? A. Yes, that was after our friends in The Hague rejected, and we rejected in the cable.

Q. Yes, but at that time no tests on the process had been made in the United States by you? A. Not at that time.

Q. Do you consider the Lefort process, if operative, to be very simple and of great potential commercial value? A. Oh, yes.

Q. And do you consider that from the chemical viewpoint the Lefort process, if it produced ethylene oxide by passing ethylene and air over silver at a temperature between 150 and 400 degrees was a radically new process industrially? A. I think as far as I know, yes.

Q. Now, what one of the Shell companies has gone into the chemicals business? A. What is that?

Q. What one of the Shell companies has gone into the chemicals business? A. The Shell Chemicals Company.

Q. When did it become active in that line, in this country? A. Oh, 1929, 1930.

Q. Since then it has been expanding its line of chemicals? A. Yes, sir.

Q. And competing with other chemical companies in this country? A. Yes, sir.

Q. Looking for new lines of products and processes? A. Yes, sir.

Q. A moment ago I asked you whether it would have been normal or obvious to a skilled chemist in 1933 to consult the literature and find how to deposit silver from silver nitrate as silver oxide, reduce to silver, on an inert carrier. I understood you to say that you do not recall whether your experimenters in Emeryville proceeded in that fashion; is that right? A. That is right.

Q. Do you know now whether if they had proceeded in that fashion they would have conducted the Lefort process, and indeed have obtained ethylene oxide formation? A. Will you repeat that question, please?

Q. Well, it is a long question; I do not want to repeat it if I can avoid it. But if they had selected an inert carrier and deposited a coating of silver on it, by starting with silver nitrate, and precipitated the silver oxide by sodium hydroxide, washing, drying, and so forth, to get the silver oxide on the supporting material, then reduced that at, say, 200 degrees, or thereabouts, Centigrade, with some hydrogen, and that then they had used that simple catalyst in the Lefort process, that is, passed over it a mixture of ethylene and air, at a temperature of 150 to 400 degrees, would they have obtained ethylene oxide? A. I do not know.

* * * [422-427].

TESTIMONY OF HENRY W. DE JONG.

Thereupon—

HENRY W. DE JONG, a witness of lawful age, produced on behalf of the defendant, having been first duly sworn, was examined and testified as follows:

Direct Examination.

By Mr. Barrows:

Q. What is your full name? A. Henry William De Jong.

Q. Mr. De Jong, where do you reside? A. Berkeley, California.

Q. Where are you employed? A. Shell Development, Emeryville.

Q. In what position? A. As a chemist.

Q. And were you so employed in the year 1933? A. I was employed in Emeryville but not as a chemist at the time.

Q. What was your position at that time? A. I was then a laboratory assistant.

Q. Under whom were you working? A. Dr. Groll.

Q. You heard Mr. Pyzel refer to the tests that were carried on in Emeryville in August, 1933? A. Yes, I did.

Q. What did you have to do with those tests? A. Well, I carried them out under the direction of Dr. Groll.

Q. You were first assistant in the laboratory in carrying out the tests? A. Yes.

Q. What papers did you have before you at the time you began the tests, if you recall? A. I had a copy of the Langedijk report No. 5205.

Q. Is this the identical paper that you had before you (exhibiting to the witness)? A. Well, it is the identical paper or a copy of it. There are no identifying marks on it.

Q. Do you recognize the papers that have been put in evidence as Defendant's Exhibits 46 and 47 as also identical papers to the paper you have before you? A. Yes, sir.

Q. What other papers do you recall having before you at that time? A. I had a copy of the French patent.

Q. And can you identify that French patent (exhibiting paper to witness)? A. Well, this is the one, it was either this or one very similar to it.

Q. What is the number of this? A. 739562.

Q. Do you recall any other papers you had before you at that time? A Dr. Groll had several other papers

with him, but exactly what they were I couldn't say. They were other patents, but——

Q. Before you started on these experiments what did you do in preparation for them? A. Well, we talked it over with Dr. Groll as to what lines we were to follow, the catalysts we were to use, and took the copy of the 5205, Dr. Langedijk's report, and looked at the method of analysis and method of operation, and also the type of apparatus used.

Q. And then did you construct a special apparatus for these experiments? A. We constructed an apparatus in accordance with the flow sheet given in 5205.

Q. That is, an apparatus similar to the apparatus which formed a copy of this report 5205, Exhibit 46? A. Yes, sir.

Q. Now, was the apparatus identical with that, or how close to it did it come? A. We used three furnaces instead of two, in order to get the right length of the catalytic space.

Q. Well, since that time have you prepared a drawing of this apparatus, recently, showing that apparatus as you recall its construction? A. Yes, sir.

Q. And is that the drawing (exhibiting to witness)? A. Yes.

Mr. Barrows: We offer this in evidence.

(Drawing of apparatus used in experimenting at Emeryville marked, "Defendant's Exhibit 69.")

Q. Now, will you describe briefly just what the apparatus was and how the process was carried out in it? A. The apparatus consisted of a tube containing three furnaces, and it had an inlet at the head of the tube to introduce the water first and a side inlet where the gas was introduced, and from the top it went into a condenser, such as a flask, to collect the materials condensed, and from there on the remaining gases passed through a series of washing bottles containing water in which they were kept in a bath where any ethylene oxide that might be formed would be absorbed, and from there the gases were led through a second trap which was contained in

a cooling mixture of alcohol and CO_2 to trap any further ethylene oxide that might be present, and as a third check they passed from there on to a bottle containing magnesium chloride which would precipitate any ethylene oxide present which was not trapped in the previous traps.

Q. Where was the catalyst located in this apparatus? A. The catalyst was located right inside of the tube for the length of these three furnaces.

Q. And how did you maintain the temperature in the tube? A. We had a thermocouple right in the center of the tube which might be moved back and forth according to the exact location of the highest temperatures.

Q. Where did you get the proportions of the gases you used in these experiments? A. The proportions were used in accordance with the report of Dr. Langedijk 5205.

Q. And you studied that report and drawing to follow it out as closely as you could in accordance with the procedure thereof? A. Yes, sir.

Q. How about the analytical tests made of the product, did you make them or were they made by somebody else? A. The analytical tests of the product were made by me, whereas the gas tests were made by the analytical laboratory.

Q. What method did you follow in the making of the test you made? A. We used a method outlined in the report 5205.

Q. And did you make records of the experiments that you made at that time? A. Yes, sir, all the records were kept in a notebook.

Q. Is this the notebook in which you made these records? A. Yes.

Q. On what pages are these experiments recorded? A. The experiments start on page 77 of the notebook and end on page 94.

Q. Between what dates were these experiments carried out? A. The dates are August 3rd, 1933 to August 31, 1933.

Q. Are all of these pages you have identified in your handwriting? A. Yes, sir.

Q. Are certain of the headings or notes in the handwriting of someone else? A. There are two headings, in the first two experiments and two notes in the first two experiments which were made by someone else.

Q. Who were they made by? A. By Mr. Hearne of our laboratory.

Q. He was working in your laboratory at that time? A. Yes, with us.

Q. But the actual carrying out of the experiments you say, and the other records were made by you? A. Yes, sir.

Q. The remainder of this notebook contains nothing relating to the production of ethylene oxide for this process? A. Nothing other.

Q. And it does contain reference to many other matters? A. Yes.

Mr. Barrows: We offer in evidence photostat copies of the title page and pages 77 to 94 of the notebook produced by the witness.

(Title page and pages 77 to 94 of notebook produced by witness De Jong marked "Defendant's Exhibit 70.")

Q. Now, will you turn to the first experiment, Mr. De Jong, and state what catalyst you used in that experiment? A. In the first experiment we used a catalyst containing silver arsenic alloy.

Q. How did you come to use that particular catalyst in the first experiment? A. We had that catalyst on hand. It had been previously used for other furnace runs.

Q. And was it a satisfactory catalyst in the other processes in which it was used? A. Yes, it was with the other processes.

Q. What results did you get when you used this catalyst in this process? A. In this process we got no ethylene oxide formed.

The Court: You say you got no ethylene oxide?

The Witness: Not with that catalyst, no, sir.

By Mr. Barrows:

Q. In carrying out this and the other experiments you say you followed the same general procedure, on gas composition as in the report which was furnished you? A. Just as closely as we possibly could.

Q. Now, in the next experiment, what page is that on in the notebook? A. That is on page 79.

Q. What catalyst did you use in the second experiment? A. Silver turnings.

Q. Was that a catalyst which had previously been used in other processes? A. Yes, we had used that before, too.

Q. And was that a satisfactory silver catalyst in the other process? A. Yes, sir.

Q. What results did you get with this experiment? A. We also got negative results on this experiment in the formation of ethylene oxide.

Q. By negative results, you mean you obtained no ethylene oxide? A. Yes, sir.

Q. What was the next experiment and what catalyst did you use? A. The next experiment was number three on page 83, where we used what we call an activated silver catalyst.

Q. Was that catalyst and its preparation described in the notebook? A. It is described on page 81 of the notebook.

Q. Was that a catalyst which you had previously used in other processes? A. It was a catalyst which Doctor Groll had previously used but I had not personally used before.

Q. Were all of these catalysts suggested by Doctor Groll? A. Yes.

Q. And you merely used the catalyst he furnished you or told you to use? A. Yes, sir.

Q. Now, the method of preparing this activated silver catalyst on page 81, where did you get the directions for that? A. Those were given me by Doctor Groll.

Q. What results did you get with this catalyst? A. With this catalyst we reported some results in traces of ethylene oxide and no results in other places.

Q. Will you explain what you mean by traces, what you regard as a trace? A. In preparing the sample we use a certain amount of hydrochloric acid which is heated with the material collected and after heating, the material is cooled to determine the amount of hydrochloric acid used, which should correspond in titration to the material. If we find a small discrepancy, we put down traces of ethylene oxide formed, although there was no certainty that there was any certain amount formed.

Q. Would what you regard as a trace be any definite amount that you could give any measure to? A. It would be worthless.

Q. What is the next catalyst you tried? A. Run No. 4 on page 85, we tried a silver copper catalyst.

Q. Is the method of preparation of that catalyst given? A. It is given on page 81.

Q. Was that a catalyst which you had used before or which had been used as far as you know? A. It was new to me but Doctor Groll gave me directions for the preparation.

Q. What results did you obtain with that catalyst? A. In this run we again recorded a trace of ethylene oxide formed at temperatures around 375 degrees.

Q. And the significance of the trace is what you just explained? A. Yes, sir.

Q. What catalyst did you try next? A. Run No. 5, page 87, we tried silver copper catalyst again.

Q. The same catalyst or a different catalyst? A. The same catalyst but this time we reduced it with hydrogen previous to running the ethylene air mixture through.

Q. What results did you obtain in this test? A. In this test we reported a trace of ethylene oxide formed.

Q. What catalyst did you try next? A. Run No. 6, page 89 we tried a catalyst which is quite complicated but the preparation is given on page 88, a catalyst containing a series of elements which we took from the French patent and includes silver, lead, nickel, tin and bismuth on pumice as a carrier.

Q. What results did you obtain with that test? A. In that test we again obtained negative results, no ethylene oxide formed.

Q. What did you do next? A. Run No. 7, page 91, we reported running a test except this time we reduced the catalyst with hydrogen passing through.

Q. What results did you obtain in that experiment? A. We obtained a trace of ethylene oxide again.

Q. Then what catalyst did you use? A. On page 8, run number eight I mean, page 93, we used a catalyst which is again complex and its preparation is given on page 92, containing various elements on pumice.

Q. Do you recall how you came to prepare that particular catalyst? A. The directions were given me by Dr. Groll.

Q. What results did you obtain with that catalyst? A. No ethylene oxide was formed with that catalyst.

Q. What was the next catalyst you used? A. Run nine, page 94, we used a catalyst known as Hopcalite, which is an oxidation catalyst.

Q. Was that prepared or did you have it on hand? A. That is prepared commercially, used in gas masks.

Q. What results did you obtain on that? A. In that catalyst we had great difficulties keeping the temperature down because we got glowing in the catalyst and the result was the formation of CO_2.

Q. Did you assist Doctor Groll in writing up the report of these tests? A. Yes, sir.

Q. Is this the report? If so, will you state in whose handwriting it is and what you had to do with it? A. It is a report in Doctor Groll's handwriting and it contains a section on catalysts which is in my handwriting.

Q. Which pages are in your handwriting and which are in Doctor Groll's? A. Page one is in Doctor Groll's handwriting and pages two and three are in my handwriting and pages four and five in Doctor Groll's handwriting.

Mr. Barrows: I offer a photostatic copy of this report in evidence.

(Photostatic copy of report of Doctor Groll and Mr. De Jong marked Defendant's Exhibit 71.)

Q. I show you, Mr. De Jong, Defendant's Exhibit 65, which is pages 42, 44, 45 and 46 of the Shell Development Company report for the period ending December 31, 1933 and ask you if you had anything to do with the preparation of that report and particularly the very beginning on page 44, paragraph No. 3? A. Yes, the report was written by Doctor Groll and given to me for correction or editing prior to publication.

Q. And is that report one which you saw shortly after it was completed? A. Yes, sir.

The Court: What was that you just referred to, is that this one?

Mr. Barrows: No.

By Mr. Barrows:

Q. One more question, Mr. De Jong. In these experiments which you carried out, how much water did you use expressed in the approximate percentage composition of the total mixture of ethylene, air and steam? A. The steam amounted to approximately 65% to 75% of the whole mixture.

Q. That is, it would be some two or three times as much as the whole volume of air and ethylene mixture used? A. Yes, sir, that is right.

Q. How did that compare with the proportion of steam used in the report which you say described the apparatus and the procedure that you followed? A. It is nearly identical. In the report, I believe 70 to 75 per cent.

Mr. Barrows: That is all.

I have other photostatic copies of these original exhibits; and I understand that is agreeable to counsel for the plaintiff and the court.

Mr. Watson: We will never object on the mere ground that they are photostatic copies.

CROSS EXAMINATION.

By Mr. Watson:

Q. How old are you? A. Pardon me.

Q. How old are you? A. Thirty.

Q. So when you undertook this work you were 23? A. Yes, sir.

Q. Now, what was your education? A. I was one year at the University.

Q. What university? A. The University of California, Berkeley.

Q. What course? A. Chemistry.

Q. You are here under subpoena? A. No, sir.

Q. Who told you to come? A. Our patent department in San Francisco.

Q. Who is paying your expenses? A. I have no idea, sir.

Q. Are you getting anything beyond your expenses? A. No, sir.

Q. Now, referring to your second test, when you used silver turnings, in what other process had you used those? A. We had used them some in our ketene experiments.

Q. In what other process did you use those silver turnings? Describe the process, please? A. Well, it is a process of passing—well, the only thing is, I don't know that I am allowed to give the process.

Mr. Watson: Well, I won't pursue the subject, if the Court please; but I submit that that certainly destroys the weight of the witness's testimony.

Q. (By Mr. Watson) How are turnings made?

Mr. Edmonds: Just a minute. We would like to have this witness explain everything of the sort to counsel that he wants to know. At the same time, he feels under some obligation to his Company not to tell things that are within the company's secret procedure, unless it is needed here.

And on this particular thing I think I can tell him that the company does not object to his answering on this; and I would like to advise him that he need feel no restraint on this point, so far as the company is concerned.

The Court: Well, what I would like to know,—that might simplify things a good deal,—is whether you used silver in the way that it is used in that model that the plaintiff had explained very fully, through Prof. Stevenson.

The Witness: Yes, sir, we have used it for the dehydration of alcohols to ketones.

The Court: You did what?

The Witness: Used it for dehydration of alcohols to ketones, passing the alcohol over the catalyst, and removing the product, in the same manner as we did in this particular experiment.

The Court: You know what I am referring to, that model back of you. Did you prepare the catalyst in a form such as that?

Mr. Edmonds: The witness was not here on Thursday and Friday of last week, when that was under discussion.

The Court: Well, he can look at it now.

Q. (By Mr. Watson) Walk over and look at it. A. I can see it from here. Do you mean that particular form of catalyst?

The Court: Well, that has been explained by the plaintiff's witness as a common form of silver catalyst that could normally be used according to the teaching of this patent.

Now, my question is—I do not know whether you accept that as a fact—but without asking you to accept

it or not, I want to know this, did you prepare something akin to that?

The Witness: May I take a look at it?

The Court: Yes.

(Model referred to examined by witness.)

The Witness (continuing): We prepared something just like that for our later experiments, but not in the first silver turnings experiment.

The Court: Not in those you have just gone over?

The Witness: Oh, yes, sir. In the later ones, where we used a combination of elements on a pumice carrier.

The Court: Well, then do I understand that before you got through making these various catalysts, and giving your report, from which you say your results were unsatisfactory, that you got only very small traces of the desired product, you used, you made up a catalyst like that?

The Witness: Yes, sir.

The Court: You made up a number of others?

The Witness: Yes, sir.

Q. (By Mr. Watson) Do you know how that catalyst was made? A. No, sir.

Q. Then, why do you say you made up one like it? A. Only from a visual point of view.

Q. Only from what? A. From looking at it.

Q. Assuming that that is a deposit of silver on an inert carrier like filtros, in any of those experiments that you testified about, did you make a catalyst of silver alone, on such an inert carrier? A. Does the question specify silver alone?

Q. Yes, sir. A. Not silver alone, no.

Q. I notice in some of your tests you used a mercury treatment? A. Yes, sir.

Q. What was that on? A. That was on silver, silver turnings.

Q. Had you read the literature on the subject of oxidation catalysts— A. I had not.

Q. —at the time you undertook this work? A. I had not, no, sir.

Q. You got your information from Dr. Groll? A. Yes, sir.

Q. Did Dr. Groll tell you that mercury was known as a violent poison for catalysts? A. No, sir.

Q. In your third experiment, I think you referred to activated silver. Was that the silver turnings? A. Yes, sir.

Q. And how was the activation? That was the activation of mercury. Just what did the treatment consist of? A. It consisted of spraying mercury on silver and allowing it to stand for a certain length of time, during which the mercury was driven off by heating.

Q. At what temperature? A. I believe that is in the notebook. (Notebook handed to witness.) 500 degrees under reduced pressure.

Q. 500 degrees Centigrade? A. Yes, sir.

Q. When you used the 500 degree temperature, did you know that in the subsequent use of the silver in your apparatus, where you were passing ethylene and air, you were to employ a temperature much lower than 500 degrees? A. As a starting temperature, yes, sir.

Q. What was the temperature used in your apparatus on the test No. 2 and the test No. 3? A. In test No. 2 we started at 375, and finished at 450 degrees Centigrade.

Q. And test No. 3? A. And test No. 3 we also started at 375 degrees, and finished at 550 degrees Centigrade.

Q. You mean that in test No. 2, where you used the silver turnings, that those turnings were actually physical turnings which had been used in another process? A. Yes, sir.

Q. Not merely silver turnings of the same kind, but some which themselves had been used in this other process? A. No, sir, they were fresh.

Q. That is what I am asking you. Now, how long did that run continue? A. Which run?

Q. Run No. 2. A. Run No. 2 was carried out from 1:40 P. M. to 3:40 P. M., two hours.

Q. What is your last answer, two hours? A. Two hours, yes, sir.

Q. And that showed that you were getting no oxidation? A. It showed that we were getting no ethylene oxide formed, as far as the test was concerned.

Q. Were you getting any oxidation of ethylene? A. No, sir, not according to our gas analysis.

Q. You were getting no carbon dioxide? A. No, sir.

Q. From that you concluded that the silver did not have an active catalytic surface? Correct? That particular batch of silver did not, yes, sir.

Q. In order to give it a more active surface, you then subjected it to the mercury pre-treatment? A. Yes, sir.

Q. Now, could you observe a change in the appearance of the silver turnings after the mercury treatment? Did the surface look different in the case of test No. 3 from the silver turnings in the case of test No. 2? A. If I followed you right, the surface looked more like a gray, a sort of grayish.

Q. And how long did this run continue? A. That run was carried out in two sections: The first part from 10:30 in the morning until 12:01 P. M., and was started again at 2:25 P. M., to 4:08 P. M.

Q. Does the note at the bottom of page 83 mean that in the first two of a total of three gas analyses, you obtained no carbon dioxide? A. Yes, sir.

Q. That means, doesn't it, that the ethylene was not reacting? A. Yes, that is quite possible.

Q. And in the third sample—what time was the third sample taken? A. The third sample was taken just before the run shut down. Probably 4:05, possibly.

Q. And at what temperature? A. A temperature of 550 degrees Centigrade.

Q. And in that case you got the .6 per cent of carbon dioxide? A. Yes, sir.

Q. When you were getting no carbon dioxide, weren't you certain that there was no oxidation of ethylene occurring? A. We did not get an analysis run until possibly a day or so after the run was shut down.

Q. Well, after you got the data on this sheet weren't you certain that in that test you were getting no oxidation of ethylene? A. Well, I would take that to be certain, yes, sir.

Q. You would what? A. In my opinion I was certain.

Q. What depth of catalyst did you employ? A. You mean the length of the tube?

Q. Yes. A. Approximately a metre long.

Q. How much of it filled with catalyst? A. The entire length of the furnace, which was the entire length of the tube, practically.

Q. Was the tube you used such that the catalyst tube was arranged horizontally as shown in your drawing? A. At a slight incline.

Q. How did you fill the catalyst tube? A. We put a small amount of glass wool as a plug at one end and then put the catalyst in and put a small plug at the other end to hold the catalyst in.

Q. Was the tube entirely filled? A. For the distance of the furnace, yes, sir.

Q. You are sure it was not a case where the catalyst was simply along the lower portion of the tube? A. No, sir, because we filled in very slowly and tapped the tube as we were filling it.

Q. And what? A. Tapped the tube.

Q. To what temperature did you heat the apparatus shown at the left end of the tube? A. That was heated to approximately 125 degrees, slightly over the boiling point of water.

Q. What was in that vessel? A. It was an empty vessel, simply an aluminum block.

Q. What did you pour into the vessel? A. Water was led into the vessel and evaporated into the tube.

Q. You mean water was put into that vessel at 125 degrees, water was dropped into it? A. No, we put it in by air pressure, from the graduated cylinder.

Q. At what pressure? A. Possibly one or two pounds pressure.

Q. Two pounds gauge, you mean? A. Yes, sir.

Q. Please explain to me, if you will, just how you put steam or water or whatever it was you used, in the vessel at the left of the catalyst tube? A. A little pressure was kept in this glass graduated cylinder which is merely for the purpose of forming the flow and the flow was controlled at this needle valve here on top, and the needle valve opened to the extent you wished water to go into the evaporator, and from here the steam is passed to the tube.

Q. I still don't understand you. You mean that the graduated cylinder at the left of the drawing was filled with water? A. Yes, sir.

Q. At room temperature? A. Yes, sir.

Q. And you put some pressure on that which forced the water through the line indicating a pipe to drip down into the vessel under which a burner is indicated? A. Yes, sir.

Q. At what rate did you feed the water? A. The water was fed approximately three cubic centimeters per minute.

Q. Where did the ethylene come from? A. Ethylene was fed in from a gas holder where it had been mixed with air previous to the time.

Q. What was the rate of that feeding? A. The rate of feed approximated three cubic feet an hour.

Q. At no time, as I understand you, was there liquid water in the vessel indicated to the left of the catalyst tube? A. No, not as far as we know. I mean the water was flashed right out of the vessel, that was the purpose of it.

Q. Did you analyze your ethylene? A. The mixture of gases at the start, you mean?

Q. I mean, did you analyze your ethylene? A. I don't quite understand your question.

Q. Are you a chemist? A. Yes, sir, but I don't know what ethylene you mean, starting or finishing ethylene?

Q. The ethylene you started with? A. You mean ethylene in the pure state before we blended it in with the air, is that the question?

Q. Did you analyze it at any time? A. Analyzed the mixture after we blended it in the gas holder.

Q. What did your analysis show? A. Well, several gas holders were made up, and No. 3 analysis showed 10½ per cent ethylene and 17.9 per cent oxygen.

Q. Did you analyze it to see whether the ethylene was pure? A. No, sir, the ethylene was bought commercially.

Q. In your ninth run where you used Hopcalite, you stated you got a glow in the catalyst? A. Yes, sir.

Q. That meant a temperature of something over 600 degrees Centigrade? A. I have no idea. We got 275 Centigrade after throwing our rate back somewhat.

Q. You are not familiar with the chart of temperatures by color? A. I have seen it once upon a time, yes, sir.

Q. What is the composition of Hopcalite? A. I have seen it but I can't recall offhand. It is a commercial preparation.

Q. Now, as to these nine experiments in how many of them did you use metal on a carrier? A. In experiments 6, 7 and 8, three experiments.

Q. And that carrier was pumice? A. Yes, sir.

Q. And in none of those did you use only silver? A. No, sir.

Q. Now, when did you enter the employ of the Shell Development Company? A. In 1930.

Q. And how long did you remain as a laboratory assistant there? A. Until 1936, I believe it is

Q. Did you ever try silver gauze as the catalyst for this operation? A. Not for this operation, no, sir.

The Court: What was the answer?

A. No, sir.

The Court: Well, isn't that one of the suggested methods of this French patent? A. I don't recall the text of the French patent.

Mr. Watson: It is, your Honor.

The Court: Let me ask you this: Why didn't you try every method suggested by the French patent?

A. Well, as Dr. Groll states in one of those reports it would lead to an endless combination of these various elements that are suggested for the catalyst.

The Court: Well, I mean more of the major methods that were suggested. They set out several of them here, don't they?

Mr. Watson: I am particularly interested in why he did not try even one of them.

The Court: As I understand it, the first patent—you had both patents before you?

A. Dr. Groll had several of the patents. I can't exactly state which ones he had before him.

The Court: Well, were you given instructions as to how to make the catalyst or were you left free to do that?

A. I was given instructions by Dr. Groll.

By Mr. Watson:

Q. The French patent you had was the second main patent, was it not? No. 739562, I think?

The Court: Well, the one I have here is 41,484.

Mr. Watson: That is one of the patents of addition, your Honor, I think.

The Court: Oh, is it? Yes, No. 739562.

By Mr. Watson:

Q. I think you have stated when you did this work you were not familiar with the literature on the subject of preparing active surface catalysts, is that right? A. Yes, sir.

Q. And you have no idea why Dr. Groll told you to prepare these in the way you did prepare them? A. As mixed catalysts, you mean?

Q. Any of them? A. Well, the mixed catalysts were prepared because we wanted the combination of the very elements that were stated in the French patent.

Q. I am asking you in the way you prepared them? A. The method? No, sir.

Q. Did you find anything in the French patents about treatment with mercury? A. I don't believe there is, no, sir.

Q. Do you know why Dr. Groll excluded from his instructions a request to deposit silver, as silver metal alone, on a support? A. No, sir.

Mr. Watson: That's all.

Mr. Edmonds: That's all.

The Court: Let me ask you one other question: You did not work directly with the patents, you did not have them before you, the French patents?

A. I had this one French patent, 739562 which I read in conjunction with Dr. Groll at the time.

The Court: 739562: And the addition?

A. No, sir, I don't recall that I saw them.

The Court: You did not have the addition?

A. Dr. Groll may have, but I did not personally have them.

The Court: You did not have them to work with?

A. No, sir, I took my instructions entirely from Dr. Groll about the operations.

Mr. Watson: And, of course, you did not have the United States Lefort before you?

A. I believe Dr. Groll had it, yes, sir.

Q. It had not been issued, so you did not have it? A. That was one written in English. I understand he had that.

By the Court:

Q. Have you ever tried to make ethylene oxide with a catalyst following the instructions of the additions to the French patent you referred to or by following the instructions of specifications in the American patent? A. Personally I have had no further work except as mentioned in the book.

The Court: That is all, sir. [429-468].

TESTIMONY OF HUGH STOTT TAYLOR.

Thereupon——

HUGH STOTT TAYLOR, a witness of lawful age, produced on behalf of the defendant, having been first duly sworn according to law, was examined and testified as follows:

Direct Examination.

By Mr. Edmonds:

Q. Doctor Taylor, will you state where you reside and what your present occupation is? A. I reside in Princeton, New Jersey. My present occupation is Professor of Chemistry and Chairman of the Chemistry Department of Princeton University.

Q. At this point, Doctor, I will have to ask you to tell us something about your education and experience and degrees and honors and so forth which would entitle you to testify with respect to catalytic operations. • • • [468-469].

* * * A. I attended Liverpool University from 1906 to 1912. I studied in Stockholm, Sweden and Germany from 1912 to 1914 and at Princeton as an instructor and other positions continuously since 1914 to the present date and since 1927 as Professor. I am the author with Professor E. K. Rideal of a textbook entitled "Catalysis in Theory and Practice," first edition, 1918, second edition, 1924, 1925 or thereabouts. I am a member of the committee on Catalysis and author of several patents in the field of Catalysis and consulting chemist for the DuPont Company for ten years in the field of catalysis and consulting chemist in the field of catalysis for several petroleum companies at the present time.

Q. I think that is probably enough. I take it you have read the original Lefort patent and also the Reissue of that patent and you think you understand the disclosures mentioned in them? A. Yes, sir.

Q. Now, will you tell us whether or not you think these two Lefort patents, the original and the Reissue, disclose the same process or different processes and, if the latter, will you set forth the basis for your views? A. It is my conclusion that the original patent and the Reissue disclosed two entirely different processes. From the language of the original specification, I gather that the original process patented by Lefort consists in the simultaneous action of oxygen and water on ethylene, permissively also in the presence of hydrogen, to produce ethylene oxide. The language of the second, the Reissue patent, differs fundamentaliy from that. As is disclosed, first of all by examination of the original filing for the Reissue in which the process claimed is for the production of ethylene oxide from ethylene and oxygen with water or steam, water vapor, and hydrogen as permissive admixtures with the two, ethylene and oxygen. The language of the first patent indicates clearly what the inventor desired to cover since he specified the types of ethylene that could be considered, ethylene from coke oven gas, ethylene from alcohol, ethylene from carbon and acetylene and if he had been living in this country, he would doubtless have suggested ethylene from the destructive distillation of petroleum. He then specified water and he described

how the water could be introduced into the reaction system. It could be introduced into the reaction system either from a steam line as live steam or it could be injected into the reaction system as water and then having named his ethylene and his water, he then suggests that for the oxygen he can use, you can make use of, for example, an air supply. He then proceeds to describe three different methods of operating this particular process and in each of those it is quite clear the process describes a process for the use of the simultaneous action of water and of air on ethylene because the first process specifies a high pressure unit into which the ethylene and air are introduced possibly under several atmospheres pressures and into which the water is to be injected by means of a pump. Now, if you calculate what pressure of the water vapor will be in equilibrium with water injected by a pump into the system at 200 degrees, it is a simple calculation to show that there will be fifteen atmospheres pressure of steam at least, and if you assume with the specification that the preferable working pressure is fifty atmospheres as it is given in the first column of the specification of the original patent, you will see that that particular thing calls for approximately thirty per cent. of steam in the reaction mixture.

Modus operandi 2 calls for the use of ethylene and air, which already has some water vapor in it—water vapor—and then in addition the claim for hydrogen, which specifies a minimum of 5% of hydrogen introduced into the system, or 5% of hydrogen—I am not quite sure whether he says a minimum of five—5% of hydrogen introduced into the system. That seems to say without the water vapor in the air, without the water vapor that is especially introduced, there is at least 5% of water vapor produced by the known interaction of the hydrogen and air introduced in the presence of a silver catalyst, and the temperatures raised.

Finally, modus operandi 3 calls for a reaction in a liquid water system, whereby ethylene is made to dissolve in the water, and the reaction is conducted in contact with the silver colloid, in contact with a silver colloid in the water system.

Now, if one goes over that particular patent, in the light of what had been previously disclosed in the literature—for example, the disclosure of Willstatter and Bommer, that the oxidation of ethylene conducted in the presence of silver, copper and osmium catalysts fails because of the oxidation of the ethylene to carbon dioxide and water—in the case of osmium, from 120 degrees upwards—in the case of copper, at somewhat higher temperatures—leading Willstatter and Bommer to the conclusion that these catalytic agencies are worthless for the oxidation process in general; knowing also, for example, that Reyerson had already disclosed that a silver catalyst spread on an inert support, such as silica gel, converted ethylene completely to carbon dioxide and water; in the light of those prior disclosures one is tempted to say that the real disclosure in the first patent is that by the utilization of water in addition to other reactants, ethylene and oxygen, one can, as I think it actually says in the language of the patent specification, one can so far suppress the oxidation to carbon dioxide and water that one can get yields of ethylene oxide.

Now, that point of view is thoroughly confirmed if one reads the original French Patent specification of March, 1931—March 1931, I think—and the several additions which are made to that patent between the time of the original convention date and its application in this country under the original convention date.

The first of the French patents has a very high degree of improbability, from the standpoint of a catalytic expert, because it specifies materials which the catalytic expert would normally suggest as unlikely, with a degree of unlikeliness amounting to almost a certainty—unlikely that they would act as oxidation catalysts. And it is only in the subsequent additions that silver is introduced into the picture. But all through the original patent, which calls for the simultaneous action of water and steam in the presence of a contact agency, the first one improbable, silver and various others introduced later, some very improbable ones also introduced later, one gets there a whole sequence of experiments regarding the progress of the French company during a period

of one year, which is finally translated into an American application, the original application.

If one goes carefully through that one finds on several occasions passages which confirm one's view that what it was intended to do was to patent the simultaneous action of water and air on ethylene.

The Witness (continuing): The original patent of the 27th of March, 1931, application date, specifies arsenic, antimony, their mixtures, and the mixtures of these latter. That is the application which I think, because of the nature of the catalysts suggested, gives one the impression that it is what is generally termed a paper patent, or, if I might express it otherwise, betrays a high degree of improbability that the process is a real one.

The first addition is to indicate new catalysts, and the catalysts are then just any mixture that can be taken, of antimony, lead, nickel, tin, zinc, arsenic. There again one has an introduction of materials that are highly improbable. Nickel especially is a very improbable oxidation catalyst introduced into the specification at that particular point, the first addition, No. 41255, date 4th of July, 1931.

In the second addition, of an application date 10th of September, 1931, one has a specific indication of the necessity of using water.

We read there, "The purpose of the present addition is to specify that the formation of glycol obtained by the oxidation of ethylene by atmospheric oxygen, in the presence of water, is necessarily derived from the saponification of the ethylene oxide obtained in the first stage by direct oxidation of the ethylene. The mechanism indicated in the principal patent and the first addition relative thereto, accordingly leads first to the production of ethylene oxide, which by a direct saponification gives the glycol. But tests made by the applicant show that the catalysts indicated do not enable one to obtain ethylene oxide by the oxidation of ethylene if the gases are dry. The presence of water is necessary for the production of ethylene oxide, even apart from any attempt to bring about saponification."

That, it seems to me, clearly indicates the intention of the authors to cover the simultaneous action of water and oxygen for the production of ethylene oxide.

The next addition, filed September 25, 1931, specifies the use of hydrogen, 5 per cent.

The fourth addition calls for various methods of activating silver by introducing small quantities of gold, copper, and iron.

Please notice, however, that the principal French patent has for its object a process of obtaining ethylene glycol, which consists principally in subjecting the ethylene to the simultaneous action of the atmospheric oxygen and water, in the presence of catalysts under atmospheric or superatmospheric pressure.

So that as far as all of the first four additions are concerned, the process is for the oxidation of ethylene in the presence of water.

That was followed by a French patent, taken out on the 3rd of October, 1931, number 739562, which bears great similarity to the initial specification, because it is noted that you obtain ethylene oxide here, whereas the original French patents had called for ethylene glycol. And, therefore, one, has in 739562, a patent which is quite similar, though it differs in minor details from the original American specification, and is characterized principally in that ethylene is subjected to the simultaneous action of atmospheric oxygen and water in the presence of a catalyst, and hydrogen.

That, I think, clearly reveals the amount of information—there are one or two additions to that: There is that first addition, the 19th of December, 1931. There is a second addition, which is much later than the things we are considering, because the second addition was asked for in June, 1934. So that 739562 and its additions were available to the authorities when they filed the application for what ultimately was issued as 1998878.

This particular patent appeared in a reissue form on May 18, 1937, the reissue having been applied for September 25, 1936.

The Witness: The original specification says, "This invention has for object a process for the production of ethylene oxide, which mainly consists in subjecting ethylene to the simultaneous action of the oxygen of the air and of water, in presence of a catalyzer, and if need be of hydrogen." It is interesting to see what the reissue calls for. The wording is very similar. "This invention has for its object a process for the production of ethylene oxide, which consists essentially in subjecting ethylene to the action of oxygen of the air, in the presence of suitable catalysts, at temperatures between about 150 and 400 degrees Centigrade. If desired, water, or hydrogen, or both may be introduced for the purposes of facilitating or accelerating the reaction, as more fully hereinafter described.

What the experts have to decide is whether on April 23, 1935, when No. 1,998,878 was published, this particular patent contemplated the reaction of ethylene and oxygen permissively in the presence or absence of water.

The Court: Now, let me see, just to interrupt you for a moment, if I understand you: you say that when that patent, the original one, 1998878 was applied for and issued it had as its obvious basis the French patent which you say made water an indispensable element? A. Yes, the original French patent 729952 with its several additions, and they had available to them also No. 739562 and one addition.

The Court: I just wanted to see if I understood you. Now, you go on from there.

Mr. Watson: I might say that your Honor and the witness are not in accord on that.

Mr. Edmonds: I think they are.

Mr. Watson: I got the impression the witness did not understand your Honor's question.

The Court: Well, I will repeat it. I followed the evolution as Professor Taylor has given it of the French patent and the gist of what he stated is that he considers that evolution clearly indicates that water was made a sine qua non of the whole business. A. Yes, sir.

The Court: And then you say that the original American patent, the United States patent, was based on that?
A. Yes.

The Court: That is, the French issue culminated in the two French patents. I just wanted to get that straight.

Now, go ahead, proceed from there and explain what happened after that in the United States Patent Office.

The Witness: An expert invited some time after April 23, 1935, to examine whether this process claimed the oxidation of ethylene by air and permissively with water would certainly examine all the available material that had been published in this field up to that time; and in doing that I came across another French patent for the oxidation of ethylene with air in the absence of water, and I found that that French patent was claimed by the same society that took out these patents, and they claimed that particular French patent as a new invention.

The Court: Now, you are referring to a patent that has not heretofore been referred to in this case?

A. Yes, sir. * * * [474-488].

* * * The Witness: This late French patent by the Societe Francaise de Catalyse Generalisee is No. 771650. It was filed on July 10, 1933, and granted July 30, 1934, before the original American patent was issued. And the first paragraph shows the striking variance from the two preceding patents. It says: "The present invention relates to the preparation of ethylene oxide by the direct oxidation of ethylene in contact with pure oxygen or air."

Now, I called your attention to the original specification for the reissue. I now wish to call your attention to the final specification for the reissue.

The Court: As it appears in the reissue patent?

A. Yes. It essentially consists in causing ethylene to combine directly with molecular oxygen: "The present invention of date 1933 relates to the preparation of ethylene oxide by the direct oxidation of ethylene in contact with pure oxygen or air," and then it goes on to explain why this is a valuable discovery.

"The reaction during oxidation requires special precautions against explosion hazards. In connection with the industrial oxidation of ethylene it was suggested as early as 1904 that water vapor be introduced into the reaction. The explosion hazards are thus caused to disappear but the oxidation products have always been aldehydes.

Now, the third paragraph——

The Court: I don't know as I understand that.

The Witness: Well, they are just saying that previously water vapor has been introduced and that previous literature prior to the literature discussed in the next paragraph indicates that you got aldehydes, formaldehyde, in all probability, in addition to the ethylene.

The Court: You mean water vapor was introduced to lower the hazards but in lowering the hazards you got poorer results, is that what you mean? A. Yes, they say you got aldehydes and that is a different result from the patent in consideration.

The Court: Well, would another way of putting it be this: that you reduced the hazard but you did not get as good results as you desired.

A. Did not get the desired results.

Mr. Watson: You got entirely different results.

The Witness: Yes, you got aldehydes. According to the statement of this paragraph you got aldehydes.

The Court: Well, then, if you take that literally, that second sentence is tantamount to saying that the previous inventions were no good?

Mr. Edmonds: This paragraph is about 1904, the disclosures of 1904.

Mr. Watson: That paragraph points out, I think, if the Court please, that the same reference in 1904 had nothing to do with this subject.

The Court: Well, if you read the last sentence of the second paragraph as being modified by the words, "as early as 1904," that is true, but it does not read that

way to me: "Explosion hazards are thus caused to disappear," "are thus caused," reading as of 1904.

Q. But the oxidation products have always been aldehydes? A. Yes, the claim here is that the previous work differentiates itself from that of the preceding French patent claims by this company in that ethylene oxide was not the product but aldehydes. The statement of their invention is in the third paragraph.

By the Court:

Q. I don't think I have made myself clear. If you take the last sentence of the second paragraph of this newly discovered French patent 771650, it would seem to say that this inventor has found out that it is a bad thing to use water because although it lowers the hazard of explosion, you did not get ethylene oxide. That is what the sentence says. A. But he qualifies it in the next paragraph.

Q. I haven't come to that. That is what he says, isn't it? A. Yes, and that, of course, is prior art, but I am not discussing prior art now.

Q. Now, but if you stopped there it would be tantamount to saying that he thought he had something which was really effective and that the prior French patents were really paper patents because they used water and did not produce results.

Mr. Watson: Is that what you mean?

The Witness: If he thought that in the second paragraph, he hastened to correct himself in the third paragraph.

By the Court:

Q. Let's pass to that? A. The third paragraph says the applicant has improved this process by bringing into play catalysts suitable for obtaining a specific reaction which gives ethylene oxide as an oxidation product and this improved process marked a considerable step forward in the technique in the production of ethylene oxide by the direct oxidation of ethylene but the necessity of vaporizing large amounts of water requires an ex-

penditure of calories which obviously contributes its share to the production cost of the ethylene oxide obtained.

Q. Now, you have to read, that doesn't make any sense to me unless you read the next paragraph? A. That third paragraph undoubtedly refers——

Q. But you have to read the next one, don't you, to have that really make sense? A. The applicant is stating in the third paragraph what is the substance of the first two French patents that deal with this subject.

Q. But he sums it up, read the next one, isn't the next one the important one? A. The next paragraph claims an entirely new process of the present invention; namely, those applied for on July 10, 1933, the present invention limits this. It has as its object a process for producing ethylene oxide by the direct oxidation of ethylene without water vapor and with no explosion hazards. In the next paragraph, the process is characterized principally in that the ethylene-oxygen or ethylene-air mixture is diluted with a large amount of inert gas and as inert gases he specifies first a particularly advantageous form, the application or use of carbon dioxide; that is one inert gas; and then when the claim is finally resumed on page 5, methods of carrying—the bottom of page 5, methods of carrying the invention into practice, it is characterized in that the inert gas used is nitrogen; the inert gas is carbon dioxide, which owing to its partial pressure action limits the harmful reaction yielding carbon dioxide and water; the oxidation takes place in the presence of a catalyst constituted by pure silver, silver activated by gold and copper. In the use of a mixture composed of ethylene, pure oxygen and an inert gas, nitrogen, you have a new invention claimed for the first time on July 10, 1933, granted by the French patent office, an English equivalent to it filed and no American equivalent of that application filed or at any rate none issued so far in this country.

Q. And you think the American applicant for the Reissue had that before him and in his mind when he made the application? A. I think he would have been very foolish if he had not.

Q. As I understand it, you go further and you think it was so clearly reflected in the Reissue statement that he must have had it before him? A. The parties at issue are identical in each case.

Q. What was that last, I didn't understand you. A. The parties involved in the three patents are the Societe Francaise de Catalyse Generalisee. There are three patents and they are all in the name of the Societe Francaise de Catalyse Generalisee and in the third French patent they cite the first two and say we have improved it.

By Mr. Edmonds:

Q. Professor, at this point it would be appropriate to refer to the three British patents. Do you have those there? A. There is a British patent 402438 bearing a convention date October 3, 1931, which is the convention date of the French patent 739562. This patent application is similar in its construction to the original American application. It calls in each case for the simultaneous action of air or oxygen and water and gives the same three modus operandi that are given in the American patent 1998878. The second British patent——

By the Court:

Q. Just a minute, this is 402438 you are talking about? A. Yes, sir.

Q. Very well. A. 402749 with a convention date of December 19, 1931, again embodies the process for the production of ethylene oxide involving the simultaneous action of oxygen and of water and four claims covering that.

By Mr. Edmonds:

Q. These two British patents, do they correspond exactly with the first two of the French patents that you have described Nos. 729952 and 739562? A. They cover them substantially but there are differences due to differences in translation or in going through the office as far as I can judge.

Q. Well, now, we come to the third British patent? A. The third British patent on the same subject is 431966

with a convention date of July 10th, 1933, corresponding to that of the French patent 771650, which I have described as the third French patent.

Mr. Watson: I don't know whether it will change your Honor's ruling but I understand the fact to be that this is not a British patent. The patent application was opposed and the patent was never granted.

Mr. Edmonds: Well, call it a publication then, it is just the same, it is a publication.

The Court: I will allow it in.

The Witness: It is a complete specification and it claims the substance of the third French patent of the 10th of July, 1933, namely, the method of producing ethylene oxide by the catalytic oxidation of ethylene in the presence of an inert gas and in the absence of water.

By Mr. Edmonds:

Q. Are you reading from this third patent? A. Yes.

Q. That is explained in greater detail on the first page, at the bottom of the first column? A. Yes, there you have the same leit motif that you have in the French patent. The present invention is based on the discovery that an inert gas can be used to replace water as a diluent in the catalytic oxidation of ethylene and is therefore differentiated from the previous proposals by the use of a substantially dry reacting mixture.

Q. What about these previous proposals? A. The previous proposals are undoubtedly the proposals embodied in the previous specifications of this company with respect to the method for the production of ethylene oxide.

Q. In other words, the two British patents you have just told us about? A. Yes.

The Court: I don't understand one thing. It was stated that this last one never matured into a patent in England. The copy I have has the same appearance as the two that are patents.

Mr. Edmonds: There are some proceedings in England whereby a patent, after being granted, may be can-

celled or revoked in some proceeding and that might have happened, but this patent was certainly issued and is here for that reason.

The Witness: It says the complete specification was accepted July 18th, 1935.

Mr. Watson: But was never sealed. Mr. Edmonds suggests a publication and I think that is sufficient for his purposes.

By Mr. Edmonds:

Q. Look at the paragraph beginning at line 65 of Patent 433916. Does that make the meaning more definite? A. Yes, it defines the present invention once more, that is, according to the present invention ethylene oxide is produced by the catalytic oxidation in the presence of an inert gas and in the absence of water. That is claimed as a new invention.

Q. And by the same company that took out the earlier two patents in France and the earlier two patents in England? A. Yes.

Q. Do you want to proceed to relate them to the Reissue of the U. S. Patent? I think you have stated that there is no patent of the U. S. corresponding to the third British or third French patent.

The Court: How is that?

By Mr. Edmonds:

Q. There is no patent of the United States, no original patent of the United States corresponding to the third British patent or the third French patent. That is, there is no U. S. patent in existence based upon this 1933 invention of the third French or the third British patents.

Mr. Watson: Mr. Edmonds stated before and we will concede that there was no such application ever filed in the United States?

Mr. Edmonds: I haven't stated that, I said none had had been published.

The Witness: No, none has been published.

Mr. Edmonds: No patent has been issued in the United States, based on this 1933 invention.

The Court: Now, let me just summarize this, if I may, by a question, Professor Taylor. As I understand you, to summarize this briefly, this evolution of these foreign patents, removed from the question of technical language, you say this, that the evolution of the French patents, ending with this third, or what you call newly discovered patent, not suggested, not cited to the Patent Office, the evolution of the British patents, through the third or last British patent, No. 431966, disclose between the time of the issuance of the original American patent in suit and the issuance of the Reissue patent, a new method which does away with the addition of water; and you say that it is the necessary reasonable presumption that that evolution of the foreign patents culminated in those two new French and British discoveries, or in those two French and British patents just referred to must have been in the mind of Lefort when he tried to accomplish, as you claim he did, not by a new American patent, but by the reissuance of the old American patent, the same thing that had been covered by new patents in France and in England.

The Witness: Yes, your Honor.

The Court: That is putting in rather poor phraseology what you have expressed. Is that correct?

The Witness: Yes. * * * [493-508].

* * * By Mr. Edmonds:

Q. Dr. Taylor, during the latter part of yesterday's session you told the Court about the first and second French patents of the French company, with their several additions and the corresponding British patents, all of 1931, and the corresponding application in the United States fo. the original Lefort patent; and then you proceeded to bring in the third French patent of 1933, and the corresponding British patent, and remarked that there was no U. S. patent corresponding to them.

Now, I wish you would proceed to bring into that discussion the reissue of the Lefort patent, and relate it to

these French and British patents that you discussed. A. I would like to summarize, first of all, the French patent, the third French patent, 771650, application date July 10, 1933, in the following form: The patent has for its object a process for the preparation of ethylene oxide by direct oxidation of ethylene without water vapor. That is to be found on page 2 of the translation. "It has for its object a process for preparing ethylene oxide by the direct oxidation of ethylene without water vapor, and with no explosion hazards."

The resume specifies that it is a process for obtaining ethylene oxide by the direct oxidation of ethylene by means of pure oxygen or air, characterized principally in that the ethylene-oxygen or ethylene-air mixture, is diluted with a large proportion of an inert gas.

And as methods of carrying the invention into practice, they suggest the methods of carrying the invention into practice are characterized (a) by the inert gas used, as nitrogen; (b) the inert gas, carbon dioxide; (c) the oxidation reaction takes place in the presence of a catalyst, constituted by pure silver, silver activated by small amounts of gold, or copper, or of both gold and copper; (d) the reaction takes place under atmospheric pressure; or (e) the reaction takes place under a pressure which may reach 50 kilograms per square centimeter. That is the equivalent of 50 atmospheres.

Now, I called attention to the fact that an American equivalent of this particular French patent has not yet been disclosed. The reissue patent was applied for reissue on September 25, 1936, some two years after the publication of this third French patent. And it is pertinent, therefore, to examine in what respect the disclosures of the French patent, of the third French patent are reflected in the changes that come into the reissue as against the original specification.

And I would like to treat these under several headings. First of all, I would like to deal with the matter of water.

The original specifies, as is known, the simultaneous action of oxygen of the air and of water in the presence of a catalyzer, whereas the file wrapper for the reissue has a very similar phraseology: "This invention has

for its object a process for the production of ethylene oxide which consists essentially in subjecting ethylene to the action of oxygen of the air in the presence of suitable catalysts at temperatures between about 150 and about 400 degrees C."

The disclosure in the original application for the reissue, and also that given in the first paragraph of the reissue as issued, calls for the reaction of ethylene combined directly with molecular oxygen in the presence of a surface catalyst, which is also the claim of the third French patent.

In other words, the claim in the first paragraph of the reissue and of the file wrapper form of that is for the reaction of two constituents instead of three: ethylene and oxygen instead of ethylene, oxygen and water. The file wrapper also discloses that water is permissive and not mandatory——

The Court: What is that? I did not understand that last.

The Witness: The file wrapper discloses——

Mr. Edmonds: The file wrapper of what?

The Witness: Of the reissue patent, the file wrapper of the reissued patent discloses that the use of water or hydrogen is now permissive, where formerly it was mandatory.

Since the utilization of water is now no longer mandatory, it follows that there is now no longer any need in the reissue to discuss or to attempt to interpret the function of the water, and, therefore, the parallel passages, that are to be found discussing the function of the water, to be found in the original patent 1,998,878, are now no longer to be found in the reissue.

In other words, since water is now not mandatory there is no longer any necessity to interpret the function of the water. That is also in agreement with French patent No. 3, since that being an invention for the interaction of ethylene and oxygen there is necessarily no discussion of the function of the water in the reaction • • • [512-518].

* * * Those, I think, represent the influences exercised by the third French patent, as I read it, on the reissue in respect of water.

Now, we come to the question of the catalyst. The original specification called for a catalyst which should catalyze a reaction between three constituents: ethylene, oxygen and water. The reissue in agreement with the third French patent specifies catalysts which shall promote the reaction between two constituents, namely, ethylene and oxygen.

Next, I wish to point out that if one reads page 1, column 1, lines 5 to 7 of the reissue, one finds there the specification of a surface catalyst in terms of results that it achieves: "In the presence of a surface catalyst which favors the oxidation of ethylene to ethylene oxide under these conditions." The conditions have been specified as about 150 to about 400 degrees Centigrade. That is a functional definition of the catalyst which shall be used, and with respect to that disclosure in the specification I feel that it will be necessary to say something more when I come to the parallel problem in the claims that are made.

The Court: I did not quite follow you when you spoke about the catalyst referred to in the reissue as relating to reaction of two elements. I mean, is there anything to indicate they are exclusive as to that? That is what I understand.

The Witness: The catalyst is specified as a catalyst which causes ethylene to combine directly with molecular oxygen, that is, a catalyst to cause the combination of two substances; whereas the first specification calls for the subjecting of ethylene to the simultaneous action of oxygen and of water in the presence of a catalyzer, and, therefore, a catalyzer for three reactants.

The Court: But it could be the same catalyzer under the specifications of both patents?

The Witness: It might be.

In further specification of the catalyst one reads on page 1 of the reissue, column 1, lines 46 and following:

"Such a surface catalyst as one composed of silver in finely divided form and which may be disposed within the tube on a supporting material or in other known ways."

Further, in column 2 it is specified the surface catalyst is preferably silver as stated but other metals such as bismuth and antimony may be used; likewise oxides of these metals or likewise a combination of these metals or their oxides may be used. Experiments have also indicated that the catalysts from silver activated by the addition of small amounts of gold, or copper or iron"——

Now, I wish particularly to call attention to this quotation from the patent specification, because this is for a catalyst to cause ethylene and oxygen to combine, which is what the third French patent calls for, and it calls for precisely the same catalyst: silver, silver activated by small amounts of gold or copper.

The bismuth and antimony and the iron are in the reissue but for a different purpose than that for which they are in the original specification. There are in the original specification a large number of catalytic agents specified, and these are catalytic agents for the simultaneous action of oxygen and of water on ethylene. Now, the catalysts are catalysts for the action of oxygen on ethylene and one presumes the same list of catalytic materials will not be as efficient in the three component system as in the two component system, and hence the number has been considerably restricted. So much for the catalyst in the specification.

I turn now to the question of heat absorbing diluents because it will be noted that the French Patent No. 771650 specifies the incorporation in the ethylene and oxygen mixture of diluents such as nitrogen and carbon dioxide.

I find on page 1, column 1, line 32, I think it is, "Oxidation of ethylene takes place with the giving off of heat, and it is, of course, desirable to maintain the temperature of the zone of reaction within the range specified. This can be facilitated by suitable dilution of the reaction gases such as that accomplished by the use of air as the source of oxygen, that is, using the nitrogen as a

diluent and some water or carbon dioxide in addition to that formed can be admitted to the mixture." That is, the use of the carbon dioxide as a diluent and as an addition water is specified as a diluent. Finally it says, "Hydrogen may similarly be added," and suggesting that hydrogen may also be used as a heat absorbing diluent, whereas my experience would lead me to suggest that it would combine rapidly with the oxygen present in the system and be converted into water, and it would be the formed water that would, if anything, act as a diluent.

Now, I find no mention in the original specification of anything concerning heat absorbing diluents, and to that extent it seems to me that the reissue has departed from the original and departed in a direction which is suggested by the third French patent 771650.

Now, I would like to turn to the corresponding items in the claims.

The Court: Before you pass to that, let me ask you one question: Did you find that there was similarly no mention of the heat absorbing diluents in the earlier French patent?

The Witness: I would have to look that up specially, but my recollection is that that is so. Now, if I might take up the claims of the Reissue with respect to these three items, claims 1 to 7 each specify three component reaction systems, as does the original, and do not appear to me to have been influenced in any way by the third French patent. With respect to claim 8 the difference is at once manifest. Claim 8 derives, so far as I can see, from the third French patent in two respects; first, the process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportion of one atom of oxygen to one molecule of ethylene. The third French patent specifies the direct oxidation of ethylene and says without water. This says the direct chemical combination of oxygen with ethylene and does not mention water. Still referring to this question of the presence or absence of water, claim 9 similarly covers the direct chemical combination of oxygen with ethylene which comprises forming a mixture containing ethylene

and molecular oxygen and a heat absorbing diluent, claiming therefore something which in the third French patent was claimed as a new invention. Now, turning back to the question of the catalyst.

By the Court:

Q. Just before you get on that, will you please let me ask you a question. Taking up the 8th and also the 9th claims, maybe you are going to discuss that in the light of the catalyst. What I had in mind, I wanted you to say something about the phrase "In the presence of water," found on the 34th line and the 26th line of the 8th and 9th claims? A. That is on line 34.

Q. In the 8th claim you will find it on line 34 and in the 9th claim you will find it on line 25? A. May I discuss that with reference to the catalyst?

Q. Yes. A. The catalyst as disclosed, as claimed, in claim 8 represents a very broad claim because it says that I shall do this reaction, with an active surface catalyst of a material having the catalytic characteristics of silver. What are these catalytic characteristics of silver? These catalytic characteristics of silver are these: Catalytic characteristics of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form what? To form oxidation products containing ethylene oxide in the presence of water.

Now, in the first place, that is a functional definition of a catalyst. In the second place, that is a definition of a catalyst of extraordinary breadth because it says that if anyone now or in the future finds a substance which will cause ethylene and molecular oxygen to form oxidation products containing ethylene oxide in the presence of water, they will come within the claims of this particular patent whether that was known to the applicant at the time of this application or not.

May I give an illustration? In 1921 Willstatter showed that ethylene and molecular oxygen could be oxidized in the presence of an osmium catalyst. He showed that the reaction began a little above 100 degrees and that it was practically complete with the consumption of all of the

oxygen at about 200 degrees and that the products were carbon dioxide and water. So far as I know nobody has ever since produced ethylene oxide by the oxidation of molecular oxygen and ethylene with an osmium catalyst. But if someone comes along and succeeds in forming osmium into such a form that the uneconomic reactions of producing carbon dioxide and water can be suppressed and the ethylene oxide becomes an important product of the reaction, oxidation products containing ethylene oxide in the presence of water, so far as I read the functional nature of this particular claim, that discovery which is not yet exhibited in the literature would be barred from application by a claim of the breadth of this particular claim. In other words, this particular claim, instead of doing as one might be led to suspect from the patent specification, restricting down the number of materials in which this particular process can be carried out, is in reality spreading out to include not only those that are known now but any such as may be found in the future to oxidize ethylene and molecular oxygen to form oxidation products containing ethylene oxide in the presence of water.

Q. In other words, you mean that when you use the words "having the catalytic characteristics of silver," that is very much broader than using the words that are in the ninth claim, "With an active surface catalyst metal comprised essentially of silver"? A. Oh, yes.

Q. In other words, one has to be essentially silver and the other does not have to be silver at all. Is that what you mean? A. That is exactly what I mean.

Q. Because you can find a lot of other metals that would have the catalytic characteristics of silver? A. So defined. These catalytic characteristics are defined, they are defined as those which favor the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water.

Q. And you say there are a large number of those? A. No, I don't say there's a large number but I say if anybody finds one not disclosed before, then he would immediately have this broad claim standing against him.

Q. I understand. A. The functional nature of the catalyst does not end unfortunately at that particular point. The functional nature of the catalyst is contained also in the following clause, not only that you produce these oxidation products but you must define your catalyst in terms of "correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide." There is a tremendous amount of correlation to be carried out but I find nothing in the specification which tells me how I shall carry out that correlation and what is the nature of the correlation that I should get. I find 150 to 400. I don't find anything about the velocity of passage.

Q. Is that true of the third French patent? A. The third French patent actually gives some specification. It gives a perfectly definite specification under the first method of operation, page three, it specifies a mixture, for example, 66% carbon dioxide, 3.4% of ethylene, 6.3% of oxygen and 24% nitrogen; a mixture of air, plus ethylene plus CO_2. It specifies the reaction temperature. It specifies the catalyst used. It specifies, it says that in tests conducted by the applicant the velocity of the gaseous mixture varied between .85 and 250 litres per hour over a catalytic distance of 60 centimeters. The tube was 63 millimeters in diameter and the reaction temperature about 375 degrees Centigrade, a quite specific correlation of the various factors that enter into the matter. The same difficulty arises in the specification of the catalyst in claim 9.

Q. I thought you said it was more restricted? A. Yes, but the same functional definition of the catalyst occurs in claim 9. It will be obvious that there was a particular difficulty arising in connection with the specification of a silver catalyst for the direct oxidation of ethylene with molecular oxygen. That difficulty arose because previously there were in the literature records of people who had reacted ethylene with oxygen in the presence of a silver catalyst, namely Willstatter and Bommer and Reyerson. In order therefore to specify the particular

type of silver, it became necessary to avoid the anticipatory disclosures containing the reaction of ethylene and oxygen in the presence of silver made previously. That has been accomplished by the functional definition of the silver catalyst.

It has been specified as a catalyst composed essentially of silver, which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen, to form oxidation products containing ethylene oxide in the presence of water, and which does not favor the existence in said products of substantial amounts, of aldehydes. That is a functional definition of silver, in terms of the results to be achieved. And in addition it is anticipated, so far as silver is concerned, by the French patent 771650, which has no American analogue, and which after October 13, 1934, was published.

Concerning, therefore, this silver catalyst, I would say that it is specified in terms of the results to be accomplished; and, secondly, those results are already claimed in an earlier French patent, 771650.

Finally, there is the same correlation defining the silver catalyst as I outlined in connection with Claim 8.

That, I think, sir, expresses my present views with regard to the influence exercised by French 771650 on the reissue patent. * * * [518-531].

The Court: * * * but I would like to ask these questions:

Professor Taylor, you have not yet gone into the question of whether it is possible to follow the teachings of the Reissue Patent and successfully produce the desired result, as one or more of the other witnesses have done. But with that in the background, I would like to see whether I understand what you have said, summarizing it very briefly.

The main point which I understand you have been endeavoring to bring out is that you feel, first, that there is a very definite difference between the original patent and the reissue, in that the original provides for the use of the three elements, including water, whereas the reissue, according to your analysis of it, leaves water out

entirely in the reaction process, the major process that is stated; and then, secondly, if I understand you correctly, you believe that that change is very definitely reflected, or very definitely reflects the third French patent, and leaving out the matter of the catalyst and the diluents, those may be said to be the two major thoughts that you now have.

The Witness: Yes, sir.

The Court: I do not mean that you do not think the others are important, but those two, if I understand you correctly, are your two major points——

The Witness: Yes, sir.

The Court: In your testimony so far. Now, taking up the first, is it your contention that this departure from the mandatory use of water is such a departure in the art as you understand it as must necessarily lead one to the conclusion that the two processes are in fact very distinct? I am not trying to get you into any legal question as to whether in law they are distinct or not, but whether in chemistry they are distinct. In other words, suppose you were told to avail yourself of the prior art method, and to proceed to manufacture ethylene oxide, would you feel that if you had to do, or had to give directions to some one to do it with these two processes, one with the use of water and one without the use, they should be treated as definite, distinct methods?

The Witness: Oh, yes, your Honor. That is just what is abundantly evident in the three French patents. In the original patent they made a process for the manufacture of ethylene oxide, using ethylene, oxygen and steam. There were various additions to that patent, representing increasing knowledge of the inventors concerning the process in question.

That was directed also in the second patent. But still the process was the interaction of three elements.

Then the French company, who had gone through this whole program of work, came along and said, we have found an entirely new process, for which we use a new invention, for which we claim very considerable advantages over the previous ones, in that we now no longer

need to put in the expensive water, and have to take it out again, but we can work entirely without water added to the reacting constituents.

And I rely upon the evidence of the French people themselves, that they had two entirely distinct inventions. And the fact that they filed a third patent specifying this indicates to me quite definitely that on or about July, 1933, they discovered that they could make a much better process than they had ever before, and they proceeded to file an application asking for protection for that process.

The Court: Well, let us assume that it is better, and let us assume that under the French law it would be entitled to patent differentiation; but what I would like to develop a little more is why you differentiate it purely as a chemical question. You say it is more advantageous. Well, we have a lot of patent matters where alternates are more advantageous; but they may be so commonly suggested as an alternate that they are not entitled to separate patents. In other words, they may be something that is suggested by the first method, and something which one skilled in the art would normally try. So the fact that it is an improvement, or better, would not necessarily answer my question.

I understand you to say that it is not only an improvement and better, but it is in the art to be considered as really an entirely different chemical method. That is what you say. You see, my difficulty is I am laboring under the difficulty of not having basic training in the art. Now, to tell me, a layman, that you can make this by leaving water out, or putting it in, does not get me very far, because I look upon water as the commonest of the substances, and something which is not expensive —you refer to that—and it is readily available; and I simply visualize something where, well, you can use water here or not use it, as you see fit.

Now, you have told me that in your opinion the very absence of water, the very fact that you say "Don't use water" is a radical change in the method. Now, it is right on that point that I do not quite get the full technical reason for it being so radical and so important a difference.

Do I make my question clear?

The Witness: Yes. The only way in which I can point out to you how radical a change it is, is that it is evident from the literature that people before, attempting to partially oxidize ethylene with gases containing molecular oxygen had succeeded in producing useless products, carbon dioxide and water. Lefort came along and said, if you will allow oxygen and molecular water to react with one another, you can get a useful product, ethylene oxide.

The same company came along at a later stage and said, we have done what other people have failed to do before; we have taken two constitutents and obtained the desired product.

It is scarcely true to say that water is a cheap commodity that can be put in and taken out at will. It has to be remembered that that water has to be raised to a working temperature—shall we say 200 degrees Centigrade, which requires heat, and, therefore, calories, and therefore expense to produce that heat. That is the advantage which is claimed for the process by the French patentees in their third specification.

The Court: You see, I can not assume that just because the French Government thought that the change, the leaving out of the water, entitled the inventor to another patent. I can not assume for the purposes of this practical construction that that answers the question. They may be right, or they may be wrong. Let us assume that the French patent, as far as France goes, is valid.

Mr. Watson: May I say on that, if the Court pleases, that in France an application is filed, and with no examination whatever granted. It is like a copyright in this country. The issue of a French patent does not mean anything in the sense of American law.

Mr. Edmonds: That is of no significance here because there is a third British patent corresponding exactly to the third French and which was examined for novelty.

Mr. Watson: And that one, however, as I told Mr. Edmonds yesterday, was not issued; that patent was never received.

The Court: Well, I can make my question practically the same assuming there had never been any French or British third patents issued, and, therefore, you can assume you can not trace the influence of any other person to the Lefort reissue, but if you find that to be the case, wherever he picked it up, whether suggested by somebody else or whether through his own brain, he evolved this method where, as you describe it, you say now it is mandatory that you leave water out, you say previously that the earlier patent said it is mandatory to put it in, at least, he says how you can do it in the reissue without any water and claims that is novelty and very much less expensive and more effective.

Now, I understand you to say, Professor Taylor, that you think, even putting the question as I have put it, leaving out the presumption that might follow from what has been done by others, and the legal effect that other Governments have attached to that, leaving all of that out, you would still say regardless of that if that was not in the picture you nevertheless feel that one who says, "Make this without water," is telling the laboratory man to do something entirely different in the art from telling him to make it with water? You stand or fall on that, do you? And I am talking again only as a practical chemist.

The Witness: Yes, sir.

The Court: Not from any legal point at all but just leaving all of that out of the question.

The Witness: And I would indicate the general truth of that particular assertion by pointing out that the process becomes a valuable process when it is possible to leave out the water.

The Court: It becomes valuable, becomes more valuable, according to your theory, does it?

A. Well, shall we say that it becomes economical.

The Court: In other words, it is the preferable method from a commercial standpoint, is that it?

A. Enormously preferable.

The Court: So that you think you have here not merely a departure, which is a definitely material one from some prior art, but a departure which results in a commercial improvement?

A. Your Honor, may I express it this way: that if the original and the reissue had come before this Court in the form of the original and a new patent covering the improvements in the manufacture of ethylene oxide by the elimination of water, I don't think that I would have been on this stand, because it would have been apparent to me that that reflected an improvement. What I point out is that improvement, which bears an international convention date of 1933 has turned up in a patent which bears an international convention date of 1931.

The Court: Will you state your first remarks again, that you do not think you would be here? I did not quite follow that.

A. If the question were the consideration of a patent for the manufacture of ethylene oxide involving three elements and the catalyst, and if it had been followed by a new patent bearing a new international convention date, for a patent for the production of ethylene oxide, from ethylene and molecular oxygen and constituting an improvement on the preceding one, then I would not be here because my principal argument that I have placed before the Court would have no longer been valid, and for that reason I would not be here.

Mr. Edmonds: You would then have a 1933 patent for a 1933 invention?

A. Yes, sir. Now, what I have found here is an invention which the French people claim is a 1933 invention appearing in an American reissue with a 1931 date.

Q. In the situation you have just been discussing, this hypothetical one, there would be two different patents, one a 1931 patent and the other a 1933 patent?
A. Yes, certainly.

Q. And what you are talking about is the extent of the improvement of that 1933 patent over the 1931? A. I should regard the 1933 as a definite improvement over the 1931.

Q. But that is no parallel to what we have here, in fact it is quite the opposite? A. Certainly.

Mr. Edmonds: And in that respect which you just mentioned, we have a 1931 patent for a 1933 invention.

The Court: Now, let me ask you one other question: the prior testimony on your side was virtually that one would have to thrash around indefinitely before hitting upon the satisfactory catalyst under these specifications and claims. I understand that you in effect intimate that you feel, if you believe that to be true, that you said that the catalyst in claim 8 is extremely broad in function and the same is substantially true of claim 9, but now I am going to ask you if you will be a little more specific: Do you agree or not with the prior witnesses who have said that they thought it was impossible to, without great labor, to hit upon the exact catalyst that would do the trick under this patent?

A. In my view, and specifically referring to claim 9, it would require a considerable amount of experimental work to perform all the correlations that are called for with a view to finding the best catalyst.

The Court: Well, is or is that not true of all work of this kind in this field? In other words, as a practical matter would you expect to find the catalyst more definitely defined?

A. Your Honor, I think this involves a question of deficiency of specification, and one can give a lot of testimony with regard to that. I can give you an example if you so desire.

The Court: Well, the prior testimony on your same side of the case has been to the effect that the specifications are far too broad and vague. Now, what I want to ask you is, again leaving out any legal requirements or what would be the legal prerequisites and doing it entirely from your professional standpoint, if you were

handed this description, if you were handed either one of these descriptions combined in claims 8 and 9 and were directed to proceed to manufacture commercial ethylene oxide, would you be brought up by that direction suddenly with the feeling: "Now, I have got to have more than this"?

A. Quite definitely, and I think I can amplify that conclusion by a question of history: at one period in the development of this case I made up my mind to make a silver catalyst to convert ethylene into ethylene oxide, just to see if I, as a skilled investigator in the field, would have a success. I made up my mind to use a catalyst support material which I was using for other purposes in my laboratory, and to support silver on that material in some way or other. The carrier that I intended to use was one that I am using daily in the laboratory, and was activated alumina, a well known carrier in the art today. The result of that experiment was, I never performed it, but it was told to me by Mr. Carter, who stated—— • • • [532-546].

The Witness: • • • I am referring to the test which is in the stenographic transcript in Volume 2, in which there is a definite statement that in a catalyst produced by impregnating activated alumina with silver the product was carbon dioxide and water.

Now, the condition is much more extraordinary than that because, let us suppose that I had taken exactly the same chemical substance, alumina, but instead of taking the form of alumina such as is found as activated alumina, I had used it in the form of high temperature sintered alumina, that which is known chemically as corundum, if I had taken that same chemical compound, Al_2O_3, and impregnated it with silver I would have obtained an active catalyst as indicated in this testimony of the stenographic transcript. Therefore, we have the situation, here are two catalysts, one of which is alumina as a support with silver on the top, the other of which is alumina as a support with silver on the top, one works efficiently to give CO_2 and water; the other gives a reasonably profitable yield of ethylene oxide. The chemical composition of the two catalysts is identical, the difference apparently lies

in the property of the catalyst in terms of the structure of the underlying support in the two cases and the nature of the influence of that underlying structure on the silver catalyst that is laid on the top.

The Court: Now, you say that the third French and British patents are sufficiently specific but that this reissue is not on this point?

The Witness: May I examine the British patent?

The Court: Maybe I asked you the same question before.

Mr. Edmonds: Well, he read you a great deal of detail which is represented in the French patent. Maybe the witness has not had occassion to consider whether the sum total of those are sufficient but it certainly is in a wholly different category from the reissue, which gives no figures at all.

A. It says the catalyst employed is pure silver and does not—it says it can be silver or silver activated by gold or copper or both gold and copper.

Q. Then you would say that that was too vague? A. I think that it would require experimentation to determine which particular silver catalyst would work, yes.

Q. Well, now, if the test that the law should apply to a specification in technical processes of this kind is whether those skilled in the art can take the description or specification and by following that accurately accomplish the desired result without having to seek elsewhere for aid to any material extent, if that is the test that the law should apply in determining whether the patent should be upheld or overthrown, I would think it would have to follow from what you said that this third French and third British patent should never have been allowed. Do I make myself clear? A. Yes, but of course, that is a legal point upon which I am not competent to express an opinion.

Q. Yes, I am not expecting you to pass upon a legal question but I do understand you now to say that you think that in so far as the catalyst question goes that neither the reissue, the British nor the French third

patents are sufficiently specific? A. Yes, but that is a subsidiary defense, isn't it, I mean inadequate disclosure?

Q. I understand it is and you think the main reason why the defense here against the reissue patent is sound is that it covers a distinctly new thing, but I have gotten into the other question of sufficiency of specification and I just want to get your views? A. Well, that would be a ground for attack, the ground of inadequacy of disclosure.

Q. However that may be, you are firmly of the conviction, as I understand it, that again leaving aside any legal question, you are firmly of the conviction that, as you understand the art, when one talks about making ethylene oxide by the use of three elements, he is talking about a distinctly different process from the one where he talks about leaving water out and using only the other two elements? A. Yes, sir.

The Court: I think I understand that.

By Mr. Edmonds:

Q. Is there something more you can tell us right along that same subject, that same topic, in this respect: Is there something about the effect of the water when it is introduced in the three component process, its effect on the catalyst itself, its effect on the reaction, its effect in requiring a difference in the catalyst itself, its effect in requiring a different correlation of the factors that enter into the catalyst? A. Well, of course, one's answer to a question of that kind must be, of course, largely speculative on the basis of one's general knowledge of things in this field and of the action of various reagents. I would say that the effect of the water probably resides in the fact that it is a partaker with the other two constituents of the sites on the surface which are available for reaction.

Mr. Barrows: The catalyst surface?

The Witness: The catalyst surface, that depending upon the ratio of ethylene and oxygen and of ethylene, oxygen and water. There will be different opportunities for different reactions to occur. In the first place, the formation of ethylene oxide or, if the oxygen is present

on the surface in very considerable amounts, the oxidation of that ethylene before it leaves the surface into undesirable products, carbon dioxide and water; and the three component system determines the relative amounts of these three reactants on the surface and the ratios of ethylene and oxide determining the nature of the products which are obtained.

By the Court:

Q. You have not specifically in your last analysis said anything about the phrase "in the presence of water," used in both of the claims, but if I understand you correctly, you mean to say that that water which is not put in at all as part of the reaction, that water is not the water that might be added as part of the original reaction but it is rather the result of what takes place, is that right? A. Yes, because it says "To form oxidation products containing ethylene oxide in the presence of water."

Q. Well, my point is just this, in layman's language, do I understand you to say that no one could reasonably in the art construe that phrase as suggesting that you started off with the addition of water? A. If it was intended to convey the idea that one ought to start off with water, why shouldn't it be in the first three or four lines?

By Mr. Edmonds:

Q. In the third French patent, the process of the third French patent——

Mr. Watson: Is that an answer or question?

The Court: He answered it by asking a question.

The Witness: I will frame it in the form of an answer. I think that refers to water produced in the reaction because if it had been intended to cover the addition of water to the reaction mixture, I would have expected to find it in the first lines of the patent claims 8 and 9.

By Mr. Edmonds:

Q. What about the process of the third French and the third British patents. Is that a process where ethy-

lene oxide is produced in the presence of water even though no water is put in? A. That is the claim of the third French and the third British patent.

By the Court:

Q. Well, now, are you or are you not somewhat omitting or overlooking the sentence which has been discussed a good deal in line, beginning at line 35, the first column of the Reissue patent "This can be facilitated by dilution of the reaction gases, such as that accomplished by the use of air as a source of oxygen, and some water or carbon dioxide in addition to that formed." In other words, some water in addition to that formed can be admitted to the mixture in the reaction zone if desired? A. As a heat absorbing diluent.

By Mr. Edmonds:

Q. That is what is to be facilitated? A. Yes.

By the Court:

Q. Put in for an entirely different purpose? A. Yes.

By Mr. Edmonds:

Q. Where did that process come into this evolution which we are talking about? A. French patent 771650.

The Court: He has already covered that. I think I understand your point. Have you finished?

By Mr. Edmonds:

Q. I was going to ask the witness one question paralleling the question you asked and referring to another one of the witnesses, namely Mr. Pyzel, who testified yesterday. I would like to ask you if you are in accord with him in the statement he made which, as he expressed it, was when you have said silver, you have said nothing? A. Anybody skilled in the catalytic art knows that you have said nothing.

Mr. Edmonds: That is all.

By the Court:

Q. Well, now, that suggests one other question, Professor. I suppose to be logical then you would have to say that on this point, this latter point that Mr. Edmonds just referred to you would have to say that if only the original U. S. Patent were before you, that although it enumerates a great range of catalytic possibilities, that nevertheless it is also too vague on the question of telling you what catalyst you should use to get the desired results? A. In my view, the patent is attackable on the basis of inadequacy of disclosure.

Q. That is, you say the patent— A. The original patent.

Q. The original patent? A. Yes.

Q. And, of course, the Reissue patent? A. Yes.

By Mr. Edmonds:

Q. But its disclosure goes much further, doesn't it, than the reissue? For instance, it includes some examples which tell you something about what to do? A. Yes.

Mr. Edmonds: That is all.

CROSS EXAMINATION.

By Mr. Watson:

Q. What you have said about the indefiniteness of the disclosure, particularly in your last remark, "when you said silver, you have said nothing." Is that, in your opinion, a peculiarity of the Lefort process or do you mean that that is a general statement that is equally applicable to all catalytic processes? A. I mean that good patent applications will specify as closely and as carefully as possible the particular method of preparation of material in order that with a minimum of labor possible somebody interested in the process can proceed to repeat the process * * * [547-558].

* * * Q. You are a patentee? A. Yes, sir.

Q. I show you patent 2168590, and ask you if that is yours (handing patent to witness). A. Yes.

Mr. Watson: I ask that this be marked for identification * * * [559].

* * * (Patent No. 2168590, entitled "Catalytic Alkylation of Aromatic Hydrocarbons," marked "Plaintiff's Exhibit No. 10 for Identification.") * * * [561].

* * * Q. Dr. Taylor, please look at that patent—and you may quote from it, the statement as to the nickel catalyst. A. I said, "The reaction producing methane was first detectable at about 150 degrees Centigrade. Thus it is quite evident that, with an active nickel catalyst, whereas at 135 degrees," so on and so and so, a lot more.

Q. That is all about the catalyst? A. Now, your Honor, I would like to put in, in the form of testimony, the fact that nickel catalysts have been prepared in active forms since 1900, or even perhaps back of that time. There are the most specific instructions possible with regard to the preparation of active nickel catalysts. Many thousands of them have been made. Many hundreds of them have appeared in the patent specifications. And I think that anybody skilled in the art knows how to make an active nickel catalyst. If they do not know, they can read my paper, where I publish the details as to how that particular nickel catalyst was made.

Q. Now, do you mean to imply that the preparation of an active silver surface catalyst was not known and stated in the literature before 1931? A. I have never seen any reference to an active silver catalyst for the oxidation of ethylene to ethylene oxide prior to this particular patent.

Q. That was not my question, and I will have to pursue it. Do you mean to imply that active surface silver catalysts were not described in the literature, and how to prepare them was not described in the literature, for oxidation processes before 1931? A. I used the silver gauze catalyst for the oxidation of methane in 1918.

Q. Well, please answer my question. A. Haven't I answered your question?

Q. No, my question is, Did the literature before 1931 contain descriptions of the preparation of an active surface silver catalyst as an oxidation catalyst? A. Yes.

Q. One of them is Benton & Elgin? A. Yes.

Q. That is a very commonly known disclosure, is it not? A. Yes.

Q. And if one would follow that procedure with silver, and then use the silver by passing ethylene and air over it, at a temperature between 150 and 400 degrees, he would conduct the Lefort process, isn't that true? A. Which Lefort process? The French patent, three?

Q. Well, he would get ethylene oxide, is that not true? A. I don't know.

Q. Did you ever try it? A. No.

Q. Have you given all your testimony of opinion here without ever trying to make a silver catalyst, passing ethylene and air— A. Yes, sir.

Q. —over it? A. Yes, sir.

Q. What oil companies were you consultant for? A. The M. W. Kellogg Company, and companies that are associated with it.

Q. What? A. The M. W. Kellogg Company, and Petroleum oil companies that are associated with it, in various researches which I——

Q. Well, is the Standard Oil of New Jersey one of those? A. I think that the Standard Oil of New Jersey is in a part of the agreement.

Q. Do you know whether the Shell Company is? A. The Shell Company may be in on some of the agreement. I don't know. My relations are with the M. W. Kellogg Company.

Q. That is a construction company, isn't it? A. Yes, sir, construction and research company.

Q. Well, research on machinery and equipment? A. Oh, no, new processes.

Q. When did you first study the original Lefort patent? A. About twelve months ago.

Q. And the reissue? A. At the same time.

Q. When did you first hear about the processes being commercially operated by passing ethylene and air over a silver catalyst? A. I was told that about a year ago.

Q. Were you informed of the demonstration conducted by plaintiff ten days ago, or two weeks ago, at South Charleston? A. Yes, sir.

Q. Did it strike you that the test as there conducted was a normal simple preparation of a silver surface catalyst? A. That is one method of preparing a silver catalyst on a support.

Q. And one method which was well known prior to 1931? A. Well, I had never heard of its application to filtros before.

Q. Well, the preparation and deposit of silver on filtros, or on carborundum or fused aluminum oxide. were all well-known things? A. I had never heard of the application of silver to filtros until last week.

Q. I am excluding filtros from my question, and saying that the process was well known for the application to a support of similar character, for example, carborundum, or fused aluminum oxide. A. I heard of that from Mr. Carter.

Q. Well, now, Dr. Taylor, haven't we all agreed that the literature before 1931 commonly stated such a procedure for preparing an active silver catalyst? A. Such a procedure? What is the procedure?

Q. What procedure do you think I am talking about? A. The procedure of putting silver on some kind of a support.

Q. Well, I am talking about the procedure which was followed at South Charleston, in plaintiff's demonstration. A. You can deposit silver on pumice that way, yes. I probably heard about that before.

Q. Or deposit it on any suitable support. A. That is a regular method of putting catalytic metals on support materials, yes.

Q. And it was well known before, generally known before 1931 as such? A. The procedure of putting catalytic metals on support materials.

Q. On support materials such as glass wool, silica fibres, fireclay, alundum, and unglazed porcelain, isn't that right? A. Yes.

Q. Since Dr. Reyerson gave his testimony in the case —I think you were not present at the time—have you read his testimony? A. Yes, sir.

Q. Anything in his expressed opinions that you take exception to? A. No. In general I agree with him.

Q. Well, there are two points of Dr. Reyerson's testimony especially that I would like to get your views on. One, you recall he referred to the statement in the reissue patent that molecular oxygen was used, and he said that to a chemist the term molecular oxygen meant that you could use pure oxygen, and therefore dry oxygen. Do you agree with that? A. Molecular oxygen is O_2, and can be dry, yes.

Q. Well, you agree that the specification of molecular oxygen to a chemist means that you can use dry oxygen? A. Surely.

Q. The other point of Dr. Reyerson's testimony on which I would like your opinion is that in answer to a question of the Court he expressed his conviction that so far as concerns the original Lefort patent, the amount of added water in the entering gases that he referred to, and that you have been referring to, was of no materiality and significance. It was that there should be some, but there was nothing critical in the amount. Do you agree with that opinion? A. Of no materiality?

Q. Yes. A. Am I expected to answer all of these questions independently?

Q. Yes, or all collectively, any way you want. A. I think it is quite evident from the statement in the patent as issued that water is a material constituent of the process.

Q. But the amount of it is not material; is that it? A. The amount of it, it would seem to me, would depend

entirely upon the conditions under which the experiment was being performed.

The Court: I would like to interject one question here. I do not want to unduly interrupt you, Mr. Watson.

Mr. Watson: Yes, sir.

The Court: But as long as I have it on my mind—I neglected to ask it before—as I understand it, Prof. Taylor, the first seven claims of the reissue patent are the same as the first seven claims in the original patent, and since all of them make water mandatory, the use of water mandatory in the process, water or steam, I gather from what you have already said in response to my various questions that to be logical you would have to say that those claims were not responsive to the specification of the reissue patent——

The Witness: Yes, sir.

The Court: —as you read those specifications.

The Witness: Yes, sir.

The Court: That is all.

Q. (By Mr. Watson) I will call your attention to the following from the transcript, beginning at page 259:

"The Court: The point made by your counsel in argument is that water is an essential part of this patent.

"The Witness: Yes."——

I will omit some words without changing the sense.

"The Court: Now, my question is, Well, then, do you mean to say that because the patent does not tell you how much to put in, it is too vague? Is that what you mean?

"The Witness: Well, I admit that it is vague; but I think it is a requirement that water must be put in.

"The Court: Do you think that can be worked out sufficiently by one skilled in the art? The application of water is a pretty common thing, isn't it, in a lot of processes. Is it necessary to a trained chemist to tell how much water he should add to the process?

"The Witness: No, I do not think it is essential at all, how much water."

Mr. Edmonds: He changed that.

Mr. Watson (Reading):

"The Court: Then, would you say that the fact that the amount of water is not specified would make any difference?

"The Witness: It is not a material difference."

Do you agree with that expressed opinion of Dr. Reyerson? A. Not in detail.

Mr. Edmonds: May it please the Court, may I ask, before we go further, that the words "at all" be changed to "to tell." The witness has told me that is what he said, and he said he was taking the question, answering your question just as you stated it.

Mr. Watson: I won't change it until I hear the evidence on it, because, if the Court please, we have had ample opportunity for that witness to take the stand and say he wanted to change his testimony.

The Court: What is the change? What does it amount to?

Mr. Edmonds: It amounts to very little.

Mr. Watson: It is changing the answer from, "No, I do not think it is essential at all how much water," and they want to change it to, "No, I do not think it is essential to tell how much water." There is more emphasis on the "at all."

Mr. Edmonds: I am merely putting it in at this time, so that if counsel want to make the change now, in talking with this witness, he will have the opportunity to do it. Mr. Reyerson, in reading the testimony told me that at that point he used the words which appear two lines above in the question, and that this is not what he said, but what he actually said——

The Court: Well, he can correct that later on.

Mr. Edmonds: Yes. He speaks as a matter of recollection of what he said, what he said in the answer.

Mr. Watson: Then the witness went on, "It is not a material difference."

Q. (By Mr. Watson) You say you agree with that? A. The teaching of the patent indicates that there are various methods of conducting the process, one of which——

Q. Let us confine ourselves, if you will, please, to the one question. Is it necessary to a trained chemist to tell how much water he should add to the process? Will you answer that? A. I should say it must be a definite quantity of water.

Mr. Edmonds: But you are permitted to complete your answer as you wish, Doctor. You were interrupted in the midst of the answer before. You can always complete your answer, if you want to.

The Witness: Well, I will try to answer it if Mr. Watson wants.

Q. (By Mr. Watson) I am reading the Court's question, the next one, "Then, would you say that the fact that the amount of water is not specified would make any difference?

"The Witness: It is not a material difference."

Do you agree with that? A. Well, you see, I have already pointed out in my own testimony that there are definite indications as to the amount of water in the three examples chosen. I have shown that in the first one——

Mr. Watson: I do not want to review your testimony; I just ask you whether you agree or disagree with the statement, first, and you may explain later.

The Witness: What is the statement?

Q. Dr. Reyerson, in response to the Court's question, reading: "Well, would you say that the fact that the amount of water is not specified would make a difference"? replied, "It is not a material difference." A. I would disagree with him.

Q. Now, you started to speak of one of your answers in your testimony on direct, where you indicated that you had referred to an amount of water which was in one of the examples of the original patent, and you calculated that at 30 per cent of water? A. Yes.

Q. Why did you assume equilibrium was necessary? A. Because I was injecting water.

Q. Well, could you not inject water which would give a water percentage of less than 30 per cent? A. Yes.

Q. You could do it anywhere from nothing to 30 per cent? A. Yes.

Q. I was somewhat surprised in your direct examination to hear you, in connection with the process involved in the case, refer to the silver on an inert carrier such as silica gel. Do you mean that silica gel is an inert carrier in this process? A. It was defined as an inert catalyst—as an inert support for the oxidation of ethylene to carbon dioxide and steam by Reyerson in his paper.

Q. I will have to repeat my question: Did you mean to give the Court the impression that silica gel in this process we are dealing with is an inert support? A. I have no idea, because I have not done the experiment myself. I should judge that it is an inert support.

Q. Then you are not familiar with the literature on this subject? A. I have read a lot of it * * * [562-577].

* * * Q. At the risk of repetition, I will have to ask you again the question: Did you ever try to use activated silver on a support or not at a temperature over the range of 150 to 400 degrees, passing ethylene and air over the silver, with or without added water entering the mixture, and fail to get ethylene oxide? A. I never did such an experiment.

Q. You never tried even that experiment, so you don't know what you would get? A. Never did.

Q. In your testimony about the original United States patent, where you have expressed the opinions we have heard on the subject of water, is it not true that we can limit our consideration on that subject of water to water in the reaction zone? A. Yes, sir.

Q. Obviously there is no role or reaction before the mixture gets to the reaction zone and none after, so what we are talking about is in the zone of the catalyst? A. Yes.

Q. I did not understand your answer to the Court's question a moment ago; you have spoken of the processes

of the original patent and the reissue patent as entirely different, and I think I quote you in referring to them as fundamentally different. Now, considering them from the mechanical viewpoint, wherein do you find they are fundamentally different? A. The fundamental difference is stated by the applicants in their two applications. In the one they claim a reaction of ethylene, oxygen and of steam in the presence of the catalyst. In the other they claim the reaction of ethylene and oxygen in the presence of a catalyst.

Q. And all your testimony has been based upon the opinion that in the original United States patent and the French patents 729 and 739, and their additions, that is, the French patents preceding the filing of the original United States patent, your testimony has been based upon the opinion that in all these ethylene, oxygen and water, that is, C_2H_4, O_2 and H_2O have been made mandatory as reactants in the process? A. That is what the specifications teach.

Q. And likewise all you have had to say with respect to the difference of the reissue patent has been premised on your opinion that in that specification and disclosure the chemical compounds of the reactants are solely C_2H_4 and O_2? A. Yes, sir.

Q. Well, referring to the process of the reissue and referring to the reaction zone, is it not true that in that case there are present in the gas mixture in the reaction zone in the presence of the silver catalyst and with the temperature between 150 and 400 degrees, C_2H_4, O_2 and H_2O?

The Witness: In the reissue you ask for?

Mr. Watson: Yes.

A. Yes.

A. In the Reissue, did you ask?

Q. Yes. A. Yes.

Q. Likewise in the original, is it not true in the reaction zone there are the substances C_2H_4, O_2 and H_2O? A. Yes.

Q. Are the original and reissue patents significantly different in your opinion in the specification of the use of silver as a catalyst in this process? A. There is only one point there where there is a significant difference. In the one case silver is specified in the examples as in the form of powder and in the form of gauze and in the reissue it is specified as finely divided silver.

Q. Is it not your opinion as a skilled chemist familiar with catalytic surfaces that the surface of an activated silver gauze is properly described as silver in finely divided form? A. I would describe silver in finely divided form as any form of silver which was produced by chemical reaction.

Q. I will ask you that same question, may we not properly refer to the active surface of silver gauze as silver in finely divided form? A. After activation?

Q. Yes. A. Yes.

Q. In other words, you did not mean to imply that the reissue patent makes any material departure from the original in that the reissue excludes silver gauze? A. No.

Q. You do not think it does exclude silver gauze? A. No.

Q. You find the original and the reissue patents alike in specifying that the reaction occurs at a controlled temperature between 150 and 400 degrees Centigrade? A. Yes.

Q. And alike in that ethylene oxide is produced? A. Yes.

Q. And alike in that ethylene oxide is produced without aldehydes? A. Yes.

Q. Well, now, coming back for a moment to your answers to my immediately prior question, let us consider the original and reissue patent disclosures of their processes with respect to the conditions present and occurring in the reaction zone which, of course, is the only place the process occurs and the only place that this suit deals with. You were told, I think, how the silver catalyst was made in the demonstration in South Charleston? A. Yes.

Q. And, of course, that catalyst produced ethylene oxide there. Now, suppose we take a silver surface catalyst thus prepared on carborundum instead of filtros, which you were not familiar with, and pass ethylene, C_2H_4, and air or O_2 and the constituents of air in which the O_2 is present, over that catalyst at this temperature of 150 to 400 degrees, will you get ethylene oxide and no aldehydes? A. I am told that you got ethylene oxide by that experiment.

Q. Except in that demonstration we used filtros and in my question I am specifying carborundum? A. In view of my conversation with Mr. Carter and the testimony he has given, I would suspect that you might get it by that experiment.

Q. That would make no difference? A. I don't know of any experiment where the particular method of incorporation on the alundum has been used, this particular method of incorporating the silver on the alundum.

Q. We understand in the demonstration apparatus filtros was used. My question was, putting it on carborundum would we always get ethylene oxide without aldehydes? A. I would expect you to.

Q. In that case, there was present in the reaction zone C_2H_4, ethylene, O_2, molecular oxygen and H_2O, were there not? A. Yes.

Q. Do you know whether there was water vapor present in the entering gas mixture before it reached the reaction zone? A. In what particular experiment and where?

Q. In the demonstration at South Charleston? A. I presume that, I am speaking from hearsay, of course.

Q. I was wondering, you may. A. I am speaking from hearsay. I understand that ethylene and air were led through two tubes and down through the catalyst.

Q. And that the air contained water vapor? A. Yes.

Q. In your opinion, would the results in the production of ethylene oxide have been substantially changed, other conditions remaining substantially alike, if the entering mixture had been dried? A. I think that there

might have been changes in the composition of the exit, yes.

Q. Do you still think there would have been ethylene oxide produced without aldehydes? A. I think that is quite probable.

Q. So that in your opinion there is a concrete example where you may prepare or where one may prepare an active surface silver catalyst on an inert support, pass ethylene and air over it and get ethylene oxide without aldehydes, whether or not the entering gas mixture was dried, is that right? A. Yes.

Q. And in both of those cases there was H_2O, water, in the reaction zone? A. Yes.

Q. So that in both of those cases, in the reaction zone there occurs a direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene? A. Yes.

Q. And that occurred in the presence of a catalyst? A. Yes.

Q. And in the presence of water vapor? A. Yes.

Q. You have had practical experience in observing the transition of industrial processes from the laboratory to large commercial plants? A. A little.

Q. You are modest? A. No, that is the truth.

Q. It is common in such transitions that a great deal of engineering as distinct from research is involved, is that not true? A. Yes.

Q. Now, you spoke as a second factor of variance between the reissue and the original, the reissue's recitation in claim 9 of a heat absorbing diluent, do you recall that? A. Yes.

Q. The substances referred to by that term "heat absorbing diluents" include nitrogen and carbon dioxide in the reissue? A. Nitrogen, carbon dioxide, water and hydrogen.

Q. Well, I am glad you bring that up. You referred in your direct examination to the specification of water

in the reissue as a heat absorbing diluent. You don't mean to give the Court the impression that a chemist would fail to know that the hydrogen when added, if desired, as stated, would burn to water? A. I would say it is common knowledge to know that hydrogen would so burn to water and therefore would not function as a heat absorbing diluent.

Q. I agree with you and I have read from the reissue patent that hydrogen was included as such, but in any event we are in agreement that a chemist reading the patent would know that hydrogen when added, if added, produced heat? A. And produced water.

Q. And heat? A. Yes.

Q. Well, now, in the original patent, on the question of variance, which is all we are at the moment concerned with, is it not true that nitrogen was contemplated? A. Contemplated in what?

Q. In the disclosure of the original patent, as one ingredient possibly of the entering gas mixture? A. Yes, it says quite definitely that the oxygen can be supplied in the form of air.

Q. In following the original disclosure in that respect, a chemist would know that the nitrogen was a heat absorbing diluent in the process, is that not right? A. Yes.

Q. And that applies also to carbon dioxide, which is also disclosed as permissive in the original, is it not? A. As a heat absorbing diluent?

Q. As a diluting ingredient of the entering gases? A. Disclosed as a possible ingredient of the reacting gases.

Q. Of the entering gases? A. Of the entering gases.

Q. Of the entering gas mixture, let's put it that way, and when so employed and if so employed under the permission, a chemist would know that the carbon dioxide in the entering gas mixture would act as a heat absorbing diluent, is that true? A. He would if he thought about it, yes.

Q. Then my notes indicate that your next topic was in claim 8, the specification of an active surface catalyst of material having the catalytic characteristics of silver and so forth. You stated, I believe, that that was a very broad, indeed, an extraordinaryly broad specification? A. Yes.

Q. Do you know any catalyst in this imaginary area of extraordinary scope which actually would fall within that specification except silver or the others mentioned? A. Osmium if it gives the results.

Q. I am asking you, Doctor Taylor, not a speculation, but so far as you know, is silver the only one that falls in that definition? A. At the present moment? Known at the present moment, do you mean?

Q. I presume you were—well, yes. A. Only silver is known at the present moment as far as I know.

Q. Silver or the other elements mentioned in the reissue patent, so then your criticism of the extraordinary scope of that language is a criticism, as I understand it, of an academic speculative possibility in the future as far as anything up to date is concerned, is that right? A. It is not an academic business. It is a very serious hindrance to future inventors.

Q. Let's limit ourselves for the moment as a practical matter up to this date, it has been no handicap, is that right? A. I don't know of anything that has been handicapped.

Q. Up to this date, as far as you know, no one has found anything but silver that will meet this specification? A. As far as I know, yes.

Q. When you speak of the factor of correlating in the claims, it is common in gas phase catalytic processes, is it not, to adjust rates of flow and proportions of the entering mixture to the particular degree of temperature involved? That is a normal part of the operation in establishing conditions which are to be maintained, is it not? I am speaking practically now, practical operation?

The Witness: One relates the velocity of conduct of gases over the catalyst in terms of the amount of cata-

lysts, the velocity of reaction, the temperature at which it takes place, the concentration of diluent, and so on, yes.

Q. That is the normal thing done always by one skilled in the art? A. Yes.

Q. And a most important requirement of it is that he shall avoid extravagant extremes in any direction, is that right? A. I presume he would do that.

Q. In response to a question of the Court, on the extent of the chemical difference between the processes of the original patent, and the reissue, as you expressed it on direct, you referred in support of your opinion to the illustration that it was evident from the literature that prior workers had not succeeded in obtaining ethylene oxide from ethylene and air, is that what you said, or is that correct? A. Yes.

Q. To whom did you refer in the literature on that? A. I referred to Willstatter and to Reyerson.

Q. You are still willing to stand or fall on the soundness of your opinions expressed in your direct on whether water makes what you have called the process of the original patent and what you have called the process of the reissue patent, chemically different? A. The French company tells me that these are two different processes, and the third French patent constitutes a new invention.

Q. All right, suppose you had never heard of the French patent, it had never been issued and no publication of the question, and suppose you didn't know anything about the earlier French patent, suppose you take just the original United States patent as granted and the reissued United States patent as granted; in your opinion are the processes of those two chemically different? A. The processes of those two are chemically different if you regard the system as a whole.

Q. Well, we have already agreed, I think, that we can confine our consideration to the ambit of the reaction zone. Thus confining our consideration, is it your opinion that those two processes are chemically different? A. Chemically different in that in the one there is a large excess of water and in the other case a very small amount of water such as produced by the reaction.

Q. There is an excess of water in both cases? A. No.

Q: Are there in both cases— A. In one case the water is added, and in the other case water is an incidental product of the reaction.

Q. And in both cases the water comes out in the exit gases? A. Yes, sir.

Q. So that in both cases there are in the reaction zone among other things C_2H_4, O_2, H_2O and C_2H_4O? A. Yes, sir. * * * [580-596].

Q. In your direct examination—I will just ask you one question here about that—you referred to activated alumina as an inert support in a test of this process which at one time you contemplated doing. A. Yes.

Q. Doesn't the literature as early as 1931 tell you that activated alumina is not an inert carrier in this process? A. An inert carrier for what?

Q. Is not an inert support, suitable— A. For what?

Q. Suitable to use as a support for the deposit of metal, silver, for the purposes of passing ethylene and air over it to obtain ethylene oxide. A. To my knowledge the literature doesn't say so.

Q. Now, on claims 8 and 9, you mean to express the opinion that if one added a small quantity of water in the entering gas mixture one would thereby cease to do what is specified within those claims 8 and 9? A. I think that if one used a mixture of ethylene and oxygen and added water you will come within the specification of the original specification. If you do not add water you would come within the patent issued to the French company, the third French patent, which covers the interaction of ethylene and oxygen in the absence of water.

Q. You have not answered the question, which I will have to repeat: Do you think that one passing a mixture of ethylene and air and some moisture over silver at a temperature of 150 to 400 degrees, to produce ethylene oxide, would be doing something excluded by the language of claims 8 and 9 of the reissue patent? A. If the water, if the moisture were deliberately added, yes.

Q. You think he would be doing something excluded by the language of those claims? A. Yes.

Q. Suppose the mixture contained 1½ per cent water, do you think that would be outside the claim? A. If that was the content of the air that would be all right.

Q. If you used dry air, but added 1½ per cent so that you had the same entire mixture, do you think that would be all right? A. You would first dry the air and then wet it again?

Q. That's right, and having thus wet it so that you still have 1½ per cent water vapor, and sending it over the same silver catalyst at the same temperature, do you think you would then be— A. You would then be really using a mixture of ethylene and air.

Q. Well, answer the question: When you are doing that do you think that you would be doing something outside the specification of claims 8 and 9 of the reissue patent? A. I think so because you would be adding water.

Q. So that you think the question of variance here boils down to this narrow compass: that if you pass a mixture of ethylene and air and 1½ per cent water vapor over a silver catalyst at a temperature of 150 to 400 degrees to produce ethylene oxide without substantial aldehyde formation, the question of whether what you are doing is within the disclosure of the first patent, as you have expressed your opinion, or the disclosure of the reissue patent, as you have expressed your opinion, is decisively dependent on whether that 1½ per cent water was water vapor normally present in the air with which you mix your ethylene, or whether that same 1½ per cent water vapor was deliberately added to dry air which you then mixed with your ethylene? Is that your position? A. My position is that if you use a mixture of ethylene and air you would be following the procedure described in the third French patent, and I would suspect that anything equivalent to that procedure would also come within the provisions of the third French patent.

Q. Well, now, we will make a little faster progress if we confine ourselves to the original United States and the reissue United States—

The Court: Now, wait, aren't you just—I think I understand what the witness has said; he has been saying this over and over again, and I think it is pretty clear to me. He has been adopting really the conditions of the French patent, and as his reasons for distinguishing it he says that the patentee, the French patentee, in the beginning of Patent 771650, says, "The applicant has improved his process for bringing into play a catalyst suitable for obtaining a specific reaction which gives ethylene oxide as the oxidation product, this improved process marked a considerable step forward in the technique of the production of ethylene oxide by the direct oxidation of ethylene, but the necessity of vaporizing large amounts of water required an expenditure of calories which, obviously, contributes its share to the production cost of the ethylene oxide obtained."

Now, I understand the present witness says that that describes the original United States patent.

Then, the application goes on, "The present invention remedies this drawback. It has as its object a process for preparing ethylene oxide by the direct oxidation of ethylene without water vapor and with no explosion hazards."

I understand the present witness to say that he interprets the reissue patent as expressing exactly the last thing that I have read, and that the fact that the water is added initially gives the distinguishing feature.

Am I interpreting you correctly?

The Witness: Yes.

Mr. Watson: Well, I have boiled that down to the question where the witness is expressing an opinion on the amount of water vapor: 1½ per cent water vapor in one case he says constitutes the process of one patent, and that same amount in the other case may constitute the other process.

The Court: No, I did not so understand.

The Witness: Your Honor, you pointed out that the first patent, as I interpret it, corresponds to the use of large quantities of water, and that the reissue patent

covers the third French patent, which deals with a mixture of ethylene and air with water absent. Now, that is what I wish my interpretation of these two patents to stand on * * * [596-603].

* * * The Court: Now, do you want to say anything more, Doctor?

The Witness: I claim that the essential distinction between the original issue and the reissue is well defined by the parallel French patents, and that the nature of the reissue is determined by the discovery that was made by the French people in 1933, to the effect that it was not necessary, as in the original specification, to add large quantities of water; it was possible to operate the process with ethylene and air, and the amounts of water vapor that would normally go with such air.

Q. (By Mr. Watson) Well, now, you certainly do not want to give the impression, do you, Dr. Taylor, that if these later French-British patents were out of existence, you would have been unable to discover any difference between the original and the reissue of the Lefort patents? A. I should have discovered that in the reissue there was something that was not in the original.

Q. And your conclusion would not have been other than you have already expressed it, that the difference made the processes entirely divergent chemically? A. Yes, in this sense, that in the one case you are injecting water with a pump, or with a live steam line; and in the other case you are allowing it to pick up the amount of water vapor that it would normally do in the course of technical operations.

Q. Now, you referred at some length many times to the French, the late French patent 771650, that had a corresponding British which was not sealed. It is not your position that the British as a disclosure is any different from the French, is it? A. It should not be, because they both have the same convention date.

The Court: I think the witness said they were the same yesterday.

Q. (By Mr. Watson) Referring to the British for a moment, 431966, according to your direct examination,

which I can not put my finger on at the moment, but I understood you to say that this disclosure of the British document, as well as of the corresponding French, is epitomized in their respective statements, that the operation thereby contemplated is one of producing ethylene oxide by the catalytic oxidation of ethylene, in the presence of an inert gas, and in the absence of water; is that right? A. It says, "It is differentiated from the previous proposals by the use of a substantially dry reacting mixture." That is what the British patent teaches.

Q. And in line 64 it says, or 65 to 70, it says: "Thus, according to the present invention, ethylene oxide is produced by the catalytic oxidation of the ethylene, in the presence of inert gases, and in the absence of water." A. Yes.

Q. And that epitomizes your understanding— A. Yes.

Q. —and the radical differentiation of this document from the earlier— A. Yes.

Q. —United States and French Lefort patents; is that right? A. And in that connection I would like to observe that 1½ per cent of water vapor, since it is present in air, could be regarded as a substantially dry mixture, no matter * * * [604-607].

* * * Q. (By Mr. Watson) One more thing: I think you referred in your direct to the Willstatter article. Was I correct in understanding you to say that Willstatter drew the conclusion that metal catalysts were utterly worthless for any purpose except oxidizing ethylene to carbon dioxide and water. A. He stated that they were utterly useless for the oxidation of ethylene to form formaldehyde.

Q. And so far as his article discloses, they were also utterly worthless to produce ethylene oxide; is that correct? A. There was no evidence in the article about that * * * [608].

* * * * * *

REDIRECT EXAMINATION.

By Mr. Edmonds:

Q. Dr. Taylor, you have examined the records of many tests of catalytic operations on the oxidation of ethylene, have you not? A. Yes, sir.

Q. Many hundreds of them? A. Yes, sir.

Q. If you yourself had performed all of the tests covered by the records that you have examined, would the time that it required run into years? A. It would have run into many years.

Q. Has anything that was said on cross here this morning caused you to want to alter anything you said on direct on the subject of silica gel, the use of silica gel as a support for a catalyst? A. I would like to make a general statement about the definition of an inert support. Silica gel, for example, is an active catalyst for the dehydration of various substances. When it is used as a material for dehydrogenation to take the hydrogen out, it is an inert material, certainly in the range specified in this particular thing. So far as I know, in oxidation processes, and certainly in the experiments conducted by Reyerson, silica gel is stated quite definitely in that paper to be an inert material in causing the oxidation of ethylene to carbon dioxide and water. Therefore, from the standpoint of an oxidation support material, support material for an oxidation catalyst, I would regard from all the evidence available to me that silica gel was an inert catalyst material, a catalyst support material.

Q. Now, I wanted to ask you this: Are you familiar with the commercial use of silver as a catalyst in a catalytic operation? A. Yes.

Q. What particular one is that? A. The conversion of methyl alcohol to formaldehyde.

Q. What is the form of silver catalyst used? A. So far as I know, it is silver gauze.

Q. And what is the form of the surface of the gauze used in that operation? A. The gauze is originally put in, is just originally put into the apparatus, and the reaction is started.

Q. It is what we might call an unactivated gauze? A. Yes.

Q. As distinguished from the activated gauze you spoke of? A. Yes.

Q. Now, one other matter I want to ask you: It has been stated in certain pretrial proceedings in this suit that there are various silver catalysts that are not effective for the purpose of oxidation of ethylene, and that there are various silver catalysts that are effective for this oxidation, in a part only of the temperature range specified in the patent. Now, I want to ask you if you agree with that? A. Yes, sir * * * [614-617].

* * * * * * *

Recross Examination.

By Mr. Watson:

Q. What temperature was used in this last catalytic process you referred to, with silver, methyl alcohol, formaldehyde? A. Well, it is hot. I don't know the actual temperature.

Q. You don't know the temperature? A. It is four or five or six hundred, I suppose.

Q. Now, the only basis for your views on the silica gel being an inert carrier, that you have just expressed, are those in the Reyerson article? A. Yes, sir * * * [618].

* * * * * * *

Thereupon——

HUGH STOTT TAYLOR (recalled), a witness heretofore produced and sworn, was recalled by the defendant, examined and testified as follows:

The Court: * * * [1155].

* * * Professor Taylor, in your original examination, as I recall it, you were very positive in your views that you thought the second or reissued American patent reflected the third French and British patents?

A. Yes.

Q. And while at that time you did not know whether or not the person that drew the claims for the reissued patent was actually relying upon those foreign patents, you pointed out it was rather significant that the language seemed to be almost parallel and almost synonymous and almost identical. A. Yes.

The Court: And then you said, if I recall it, you considered those later French and British patents to be materially different, in that they purported to dispense with the addition of water?

A. Yes, your Honor.

The Court: And that, therefore, when, as I understand, you found the same process described in the reissued American patent you reached what would be the necessary logical conclusion, from your other findings: that the reissue was a departure from the original American patent in that the addition of water was no longer made material.

Now, you have since heard the other witnesses who have told about that, first Dr. Frazer said as to the addition of the water he did not think there was any material difference between the two processes. Has anything been stated to alter your original views?

A. No, I am completely unregenerate, your Honor. I would go farther than your Honor's statement that you just made, in this direction: that I would say that in my mind the first and second French patents and the United States equivalent, offer a process involving ethylene, air, and a large quantity of water, not an addition from zero to 4 per cent, such as Professor Frazer discussed yesterday afternoon. From that point of view I am quite unimpressed with the experiments that Professor Lamb conducted. Professor Lamb, in my view, investigated the interaction of ethylene and air on a silver catalyst, whereas it is my view that the first United States patent and the corresponding first and second French patents and the British equivalents call for the reaction of ethylene, oxygen and water, and no better proof of that that I can see can be adduced than the actual language of Example 1 of the original U. S. patent, where he says at page 2, column one, lines 20 and 21, "The reaction will take place in this tube between ethylene, the oxygen of air, and water". Now, professor Lamb has really investigated the reaction between ethylene and the oxygen of the air with a little amount of water which he says got into the reaction system by accident because he thought there might be a little alkali in the air

coming in and so to take it out he actually put in a little wash bottle containing sulphuric acid to simultaneously introduce a small amount of water. Now, your Honor, there has already been offered in evidence in this case the series of experiments conducted by Mr. DeJong of the Shell Laboratories in Emeryville, California, which have a very direct bearing on the quantity of water that was in the minds of the French company in about 1933 since these experiments were inspired by contact between the—— * * * [1155-1159].

* * * The Witness: Your Honor, Mr. DeJong testified these experiments were based on report No. 5205, I think it is, which recorded the contact between the Royal Dutch Shell and the Societe Francaise de Catalyse Generalisee. Now, in these experiments as much as 60% of water was actually the normal steam content of the reacting mixtures and with that in view I would like once more to refer to this French patent No. 3, to call your attention to what the nature of the change was. Let us assume in analyzing that third French patent that it was the state of the art as understood by the French company that the reaction was between ethylene and air and about sixty to seventy to eighty per cent. of steam. What is the nature of the substitution that they propose?

The Court: Which patent are you referring to?

The Witness: I am referring to the third French patent No. 771650, the first paragraph says that it is now "the direct oxidation of ethylene in contact with pure oxygen or air." The second paragraph states that steam has been used since 1904 for purposes of avoiding explosion hazards, but that aldehydes were produced. The third paragraph says the applicant improved this process using steam and he is undoubtedly referring to his first and second French patent because he now uses a catalyst to produce a satisfactory reaction giving not aldehydes but ethylene oxide as an oxidation product. He then says or points out the drawback of that, he says "the necessity of vaporizing large amounts of water"—now, if it were a question of from zero to 1, 2, 3% of water, it would be no question of vaporizing large amounts of water. Professor Lamb had no difficulty in vaporizing such amounts of water in the experiment which he car-

ried out. He just put the water in his gas line and thereby introduced a small amount of water. It says while that large amount of water constitutes a drawback, he says it requires quantities of heat, calories, "which obviously contributes its share to the production cost of the ethylene oxide obtained. The present invention remedies this drawback. It has as its object a process for preparing ethylene oxide by the direct oxidation of ethylene without water vapor and with no explosion hazards." Now, how does it do that? I think it is significant to point out that it does that by the substitution of diluent gas and it is quite specific as to the quantity of diluent gas that it substitutes. Actually the example that it cites is a mixture made up—and I now read from the third page, the middle of the third page of this specification "A mixture is made up containing for example 66% CO_2, 3.4% ethylene, 6.3% oxygen, and 24% nitrogen." In other words, the mixture of air and ethylene and CO_2, how much CO_2, 60% CO_2, to replace the 60% of steam which we have definite evidence in this Court was what the practice of the French company was as indicated by the information that they yielded, on the basis of which Mr. DeJong's experiments were carried out.

The Court: Well, now, aren't we giving too much emphasis to the French patent? Take the Reissue patent itself on lines 39, 40 and 41. It certainly provides for some water or carbon dioxide in addition to that volume which can be assumed to be in the mixture; in other words, there we have that permission granted in the specification.

The Witness: But in reply to that, I would then just go back to the very beginning of the two specifications. The first specification begins: "This invention has for object a process for the production of ethylene oxide which mainly consists in subjecting ethylene to the simultaneous action of the oxygen of air and of water, in presence of a catalyst," and if need be in order to get some more water, hydrogen, and the Reissue says, "This invention provides a specific and novel process for making ethylene oxide. It consists essentially in causing ethylene to combine directly with molecular oxygen at temperatures of about 150 to about 400 degrees in the presence

of a surface catalyst which favors the oxidation of ethylene to ethylene oxide under these conditions," and then, "the process can be carried out at atmospheric pressure and it may also be conducted at pressures above or below atmospheric pressure." Nothing about water.

The Court: As a practical matter though, we have here a man who provided first for adding some water in indefinite amounts, depending on how he wants to do it, then we have the same man saying that he wants to broaden the specification and he says you can add some water if you want to and he still talks about this reaction taking place in the presence of water. Now, no one in this case has produced any experiment or the results of any experiment or any laboratory work which indicates that you can successfully complete this process and get the ethylene oxide if the process is kept, if the reaction is kept completely dry. In other words, Doctor Lamb was given—and by the way, in your reference to what he said he did, we must bear in mind that he was only given the one patent.

The Witness: Yes, the first patent. That was the one I quoted from.

The Court: Am I correct in that?

Mr. Watson: That is correct, only the original United States Patent.

The Witness: I quoted from that as to what the reaction was.

By the Court:

Q. Yes. Now, isn't it a fact that no one has demonstrated in this case that you can successfully make this with complete dryness? A. Yes, nobody.

Q. There is no evidence of that? A. No.

Q. If you couple with that the fact that you start off with a man saying you must have some water and you have evidence that if you have some water, if you add a little, you get it from the air or no matter how you have it, it is there, you have successful operation, and that man says "I will still allow water to be used, you can

add some if you want to or not." As a practical chemical matter, why would we say that that man is departing from his original invention when he simply refers to the fact that water appears to be less necessary, although we have no evidence whatsoever here, though it may come out to some extent. That is the point I don't understand as a practical matter. You have the first patent saying "Put some water in." The second patent says, "You can put some water in if you want," and all of the evidence in this case indicates that the successful operations have been made under conditions where there was some water. Now, why should the Court be asked to say that the man is departing from his original invention when he simply says I think you do not need to use it, you don't really need to add water like I said you did in my first patent. A. I don't know legally whether I am right, but my point of view is—and I think I expressed it in my direct testimony—that what has happened has happened and so he has obtained a 1931 date for a 1933 invention * * * [1160-1166].

* * * Mr. Edmonds: Then, may I ask Professor Taylor a question or two with reference to the excluded exhibits, this being for the record as to excluded testimony.

Q. (By Mr. Edmonds) All I want to ask you is whether or not you have examined Exhibits 66 and 67? And if so, whether or not those exhibits corroborate what you have stated about the papers that Mr. DeJong examined with respect to the amount of moisture in the air admitted to the reaction chamber, as advocated by the French company in the winter of 1933-4.

The Court: Are these the excluded exhibits?

Mr. Watson: These are the excluded exhibits.

The Court: Well, he can not testify to those. They have been excluded.

Mr. Edmonds: I ask your Honor to record on the record this statement by him, under Rule 43, as testimony with reference to an excluded exhibit * * * [1170].

* * * A. I have examined these exhibits, and read them through.

Q. And will you answer the rest of the question? Do they corroborate, do they give you the same opinion as what you said was contained in the other exhibits * * * [1171]?

* * * The Witness: I have read both of those exhibits, and I think that they do corroborate the evidence that has been given by Mr. DeJong. * * * [1172].

* * * Cross Examination.

By Mr. Watson:

Q. Dr. Taylor, you recall the language of Resume 5 of the first French Patent? A. Yes, sir.

Q. That states a chemical fact, doesn't it? A. Yes, sir.

Q. And the facts of chemistry have not changed since 1931, have they? A. No, sir * * * [1173].

* * * Redirect Examination.

By Mr. Edmonds:

Q. And do you regard that statement as contradicted by later statements? A. I certainly do regard it as contradicted by later statements. And I am of the opinion that it applies to a process which is not in issue, namely, the catalytic action of ethylene, air and water, in the presence of antimony, arsenic, and bismuth * * * [1173-1174].

* * * * * *

Thereupon——

HUGH STOTT TAYLOR (recalled), a witness heretofore produced and sworn, was again recalled by the defendant, examined and testified as follows:

Direct Examination.

By Mr. Edmonds:

Q. Doctor Taylor, will you address yourself to the state of the knowledge on this subject as it existed

prior to March of 1931 and whether or not you find in the published literature prior to that date the equivalent of what you find to be disclosed in the Reissue patent in suit? A. I think I can do that. I think I can do that very briefly. The discovery or the suggestion of the partial oxidation of ethylene to ethylene oxide is an old concept. It dates from 1862, when Wurtz suggested and tried it * * * [1194-1195].

* * * The Court: When you say suggested——

The Witness: He actually tried to do it.

By the Court:

Q. He did not do it? A. No * * * [1195-1196].

* * * The Witness: Following that I would come down to 1905 and there I would cite British patent 21941 of 1905 to Doctor Walter of Germany, in which he discusses the transformation of organic substances by partial combustion, and in which in instance 4 on page 3 of the patent he discusses the oxidation of ethylene with oxygen by passing these gases through a tube heated to 400 degrees and in the presence of catalysts as specified on page 4. "The following may be employed as such contact substances: The metals platinum, iridium, palladium, silver, uranium, vanadium, iron, cobalt, nickel, cerium, thorium," and so forth.

Q. What is the date of that? A. 1905.

Q. Is that the issuance or the application? A. That was the issuance, Anno Domini 1905, but the claim for the patent was the 27th of October, 1904.

Q. Well, now, can you summarize again, do you claim that is directly in anticipation? A. I claim that is evidence that people had attempted to produce partial oxidation products of ethylene and oxygen, of which Wurtz in 1862 had said ethylene oxide was one.

Q. Could you follow, could you take the teachings of that patent and make ethylene oxide? A. I could now, certainly.

Q. And it was not done then? A. No, he was not trying to make ethylene oxide.

Q. What was he trying to make? A. Formaldehyde.

Mr. Watson: And he made it?

The Witness: And he made it.

By the Court:

Q. You are not claiming, are you, that that is a direct anticipation? You are giving me that as a second step in the evolution? A. I am giving it as evidence that the process of passing ethylene and air over a catalyst to produce partially oxidized materials was common knowledge in 1905.

Q. But no one has yet found out what they got by doing it? A. Yes, they found out they got formaldehyde and they also got carbon dioxide and water.

Q. They hadn't found out yet what we are talking about? A. Not yet. Then I jump over to 1921 and I find there an article by Willstatter and Bommer, which is also concerned with the preparation of formaldehyde from ethylene.

Q. What is that, what date? A. The paper is published in——

Q. 1924, did you say? A. 1921, volume 422, page 36, 1921.

Mr. Fairbank: That was cited by the patent office against Lefort, wasn't it?

The Witness: That was one of the patents cited against Lefort by the patent office. Now, in that particular article it is stated that "For the oxidation of ethylene to formaldehyde, accelerators, of all kinds, for example metals such as silver or copper, as well as metal oxides" are useless. He points out that the reaction starts at 130 degrees with the formation of carbon dioxide, and at about 200 degrees centigrade the oxygen is used up. With copper and silver no formaldehyde is formed and even with an excess of ethylene no carbon monoxide appears. So he points out that with copper there is considerable reaction at 250°C, and at 400 centigrade, with a mixture of 80% ethylene and 20% oxygen, half of the oxygen was con-

sumed, forming carbon dioxide. In other words, he showed that on these catalysts carbon dioxide was formed from ethylene and oxygen. Now, I would skip to 1928 and there cite an article by Reyerson and Swearingen that has already been discussed in this case.

The Court: By whom?

The Witness: Professor Reyerson and Swearingen, an article which appeared in the Journal of the American Chemical Society Volume 50, page 2872, 1928, and in that article Reyerson passes oxygen and ethylene over silver catalysts such as silver on silica gel, and he says that he is studying the action of the silver because he suggests on page 2878 that if you were not doing that, the silica gel itself would be an active catalyst. Now, then——

The Court: Do you claim that is a direct anticipation?

The Witness: No, it is not direct anticipation because Reyerson got carbon dioxide and water. Now, the points I wish to raise with regard to the prior art are two. With reference to that background as I have given it to you, what was disclosed in the Lefort patent that Willstatter and Bommer and Reyerson had not already done? They had passed ethylene and oxygen over a silver catalyst and had not observed any ethylene oxide. What did the patent teach which these men had not done? I cannot find anything that the patent teaches that these men had not done, and, if that is true, I would suggest that the patent therefore is insufficient in its disclosure by reason of that prior art. There is another alternative. The other alternative is that you can produce ethylene oxide with silver and that the thing for Reyerson to have done was to have chosen another catalyst, another silver catalyst. If that is the solution of the problem, then there was no act of invention, on the part of the inventor. If all that it was necessary to do was to take Reyerson's experiment and use another catalyst, such as the one that Benton and Elgin had suggested in the literature prior to that time, doing exactly what he did in the region from 200 to 300, which is the region that Reyerson explored, and

using another catalyst get the result, then it seems to me there is no act of invention disclosed by Lefort in his application. That is all I have to say with regard to that aspect of the matter. It leaves us in the position where there do not appear to be any adequate directions in the patent specification over and above what those gentlemen have done before to produce ethylene oxide and, if so, the patent is inadequate in its disclosure, and it does not represent any act of invention. Then, I would think that very considerable weight ought to be given to two prior patents, two prior United States patents, the McElroy patent number 1253617, filed in 1912, renewed in March, 1916, and patented in January, 1918, and another patent to McElroy, No. 1308797, application filed on June 26, 1917, patented July 8th, 1919. Now, your Honor, I don't think that I can explain this particular patent any better than Mr. McElroy did himself and I would therefore like to read you about two paragraphs of the appropriate specification and leave the matter in that position without comment. Mr. McElroy says——

The Court: Which patent are you reading from?

The Witness: 1253617. Similar passages will be found in the second patent, but patent 1253617, at page 1, column 1, line 24, states "There are various oxygen-containing or oxidized derivatives of ethylene, such as glycol, the chlorhydrin, glycol esters, glycol ethers, ethylene oxid, etc., having properties which would render them useful in various arts if such derivatives could be produced by a method sufficiently simple, cheap and ready to make them commercially available. At present however, these bodies are not available for technical purposes being merely laboratory curiosities, made when wanted for scientific purposes by indirect, cumbrous and wasteful methods."

I now turn to the paragraph in that same column, beginning at line 50 "It is an object of the present invention to obtain these potentially valuable oxidized bodies from ethylene and other olefinic gases by a direct limited oxidation under controlled conditions.

Ethylene which is typical of the other olefinic gases is a readily oxidizable substance but being endothermi-

cally constituted, oxidation is attended with the liberation of much energy and is apt to be too far going, while the direct products of its limited oxidation, ethylene oxid, glycol and chlorhydrin are themselves also readily oxidizable. Ethylene oxid, which is the first product of oxidation, is particularly sensitive; glycol is more resistive."

"I have found however that under suitable conditions the oxidation of ethylene can be limited and regulated to the desired degree and that such a limited oxidation of the ethylene itself, is, so to speak, preferential over the further oxidation of these proximate oxidation products; that is, so long as unattacked ethylene remains in the sphere of reaction the oxidation merely goes to the first stage."

May I call attention to line 62 in this same column "Ethylene oxid, which is the first product of oxidation, is particularly sensitive; glycol is more resistive.

"In the present invention therefore conditions are so controlled as to restrain the violence of reaction, the heat of reaction being absorbed or removed as fast as evolved so as to maintain a predetermined temperature range; and an excess of ethylene is maintained in the sphere of reaction throughout the course of such reaction. While other methods of temperature control may be, and usefully are, employed dilution of the reacting bodies is desirable as this also aids in controlling the progress of the reaction. As stated, ethylene is endothermic and tends to enter into reaction with accelerating velocity; and dilution is useful in damping down the violence of the action."

Now, comes the point which I wish specifically to call attention to. Beginning with line 93 "Dilution is useful in the limited oxidation of ethylene in the gaseous state," and here follow two methods of doing that, first, "By chlorin and steam, steam being best present in ample amount." That is one method of oxidizing ethylene by chlorin and steam. It does not come into this picture at all. The second method, I quote "And by oxygen in the presence or absence of catalytics such as platinum black,

labile oxids, nitrous vapors, etc." This is the third clause of this particular paragraph. He then points out another method whereby dilution may be accomplished and he writes "And in the treatment of ethylene dissolved in various liquids; advantageously an aqueous liquid," and with regard to this third method of operation he cites on page 2, beginning line 34 and ending with line 45 a description of the introduction of ethylene itself into a liquid in which is present adsorptive material such as charcoal, coke, bone-black, platinum black, etc.," and he adds on line 40 of page two, "Such bodies may, and usually do, superadd a useful catalytic effect in aiding oxidation. For example, ethylene and air may be supplied to a hot liquid holding platinum black or colloidal palladium or the like in suspension."

I think that disclosure ought to be considered in connection with the third patent of Lefort.

The Court: Well, now, in what claims of this patent does he speak about the catalytic process?

The Witness: He has a broader claim than that, your Honor. Claim 1 is "the process of preparing useful products which comprises submitting an unsaturated gaseous hydrocarbon to limited oxidation, such hydrocarbon being maintained in excess in the sphere of reaction during such oxidation."

The Court: Was this cited to the Patent Office?

The Witness: No.

The Court: Well, are you claiming this is a direct anticipation?

The Witness: That is how I would look at it, your Honor.

Q. (By Mr. Edmonds) Is there any other claim of that patent to which you think reference might be made in connection with this matter? A. Yes, claim 31: "In the manufacture of useful products, the process which comprises submitting an unsaturated hydrocarbon to limited oxidation, and during said oxidation and while supplying hydrocarbon to the reaction chamber continuously maintaining therein oxidizing conditions."

Q. Now, most of the other claims of that particular patent are limited to this other form that you spoke of, namely, the electrolytic one? A. Yes, sir.

Q. Now, will you turn to the other patent, the other of the two McElroy patents, and state what the fact is about that as to its disclosure, and refer to any claim there which is directed specifically to the catalytic operation? A. Well, if one reads the first page of United States patent No. 1308797, one finds similar material to that which I have read at length in the first patent. And, therefore, I do not think it is desirable to repeat that reading.

I then turn to page 6 of that patent, and I find in the process of producing oxidation products of the olefines, a step which consists in subjecting the olefine——

The Court: What line are you reading from?

Mr. Edmonds: The sixth page.

The Witness: This is page 6 of the second patent, 1308797.

The Court: The last claim?

The Witness: Yes, sir. "In the process of producing oxidation products of the olefins, the step which consists in subjecting an olefin to the action of an oxidizing agent in the presence of a catalyst while maintaining the olefin in excess."

The Court: What is an olefin?

The Witness: Ethylene is the first of the series of olefins. Olefin is the generic name for a series of gases, of which the first one is ethylene.

The Court: Do you think that is an anticipation?

The Witness: Yes, sir.

The Court: Although there is no mention anywhere of a silver catalyst in this or the preceding patent.

The Witness: There is mention of labile oxides, your Honor.

Q. (By Mr. Edmonds) And what is the significance of that? A. The significance of labile oxides is that it includes silver oxide.

The Court: Do the drawings disclose a catalyst?

The Witness: No, because the patent itself devotes its attention largely to an electrolytic method of producing oxygen.

The Court: What is the difference between 1308797 and 1253617.

The Witness: They are similar in content, but they are drawn with a skill which I think requires more than a chemist to understand the difference.

Q. (By Mr. Edmonds) The difference lies in the claims, does it not? A. Yes, surely; obviously it lies in the claims.

Q. The same disclosure in both? A. You see, one says the process of preparing useful limited oxidation products, and the other says the process of producing glycols.

The Court: These were not cited to the Patent Office?

The Witness: No, sir. May I say a word with regard to the term "labile oxides" suggested as a catalyst? The term "labile oxides," in my view, includes, among other oxides, silver oxide.

Q. (By Mr. Edmonds) And is silver oxide referred to in the specification of the Lefort Reissue? A. Yes, sir.

Q. And was the term labile oxide understood for many years prior to 1931 to specify silver oxide, among other things? A. Yes.

The Court: You mean to include or to——

The Witness: I would include silver oxide among the list of labile oxides. If your Honor pleases, I can give a very definite proof that that is not an ex parte statement on my part.

Q. (By Mr. Edmonds) Well, will you proceed to do that?

The Court: What does labile mean?

The Witness: Labile means it is an oxide which fluctuates between different stages of oxidation.

Q. (By Mr. Edmonds) It may take up and give off some of this oxygen, take up more and give up some? A. Yes.

I cite from an article which I wrote in the Journal of the Franklin Institute in 1922, Volume 194, page 7. "Thus, for example, it may be seen that as catalysts for oxidation processes, oxides are an important class of catalysts."

Mr. Watson: The page?

Mr. Edmonds: Page 7.

The Witness: Page 7. I quote further: "The oxides of well-defined stability are conspicuously absent. The oxides of the alkali metals, to a less extent those of the alkaline earths, definite irreducible oxides such as magnesium oxide and aluminum oxide are not frequently to be found among the oxide catalysts of oxidation. On the other hand, the labile oxides such as those of iron, copper, manganese, silver, vanadium, tungsten, molybdenum, uranium, chromium, cobalt and cerium are frequently to be noted in a systematic study of such oxide catalysts of oxidation."

Mr. Edmonds: Does your Honor want any further elucidation of the portion of the McElroy specification that the witness quoted as, for instance, the reference there to the reaction being too far going, or the reference to the fact that ethylene oxide is the first product of the reaction?

The Court: Well, I would like to ask Prof. Taylor this: Do I understand you to say that just because reference is made to the use of silver as a catalyst in the oxidation of ethylene, although there is no claim in either of these patents, as I understand it, or at least there is no claim in the first one, I understand you to say there is no claim in the second one, for using—no, in the first one—I will take that back—you say in the first one there is no claim for using that method, nevertheless you think this is an anticipation, because the inventor refers to that as a method.

The Witness: The same disclosure is made in patent 1308797, beginning line 75 on the first page, in which claim No. 17 specifically refers to oxidation of ethylene with a catalyst.

The Court: Well, you think that the reference to it in 1253617 is enough, do you?

The Witness: Yes, because there is a claim which is very much broader than the catalytic claim.

The Court: And that is?

The Witness: Claim 1.

The Court: Claim 1.

The Witness: Claim 2, claim 3, claim 31.

The Court: Well, do you claim that you could have taken either one of these patents and successfully, by following their teachings, have made ethylene oxide with a silver catalyst?

The Witness: I think it is abundantly evident to me that I could.

The Court: Well, could you at that time have done it, at the time of these McElroy patents, without the temperature specifications and the other specifications that are contained in the patents in suit?

The Witness: Yes, because he says that, "I must predetermine my temperature range so as to maintain a predetermined temperature range," lines 79 to 81.

Q. (By Mr. Edmonds) To you does that sufficiently specify an elevated temperature? A. Yes.

The Court: Where is that?

The Witness: Because he says——

The Court: What page now are you reading from?

The Witness: I am reading on the first page.

The Court: Of 1380797?

The Witness: I will read from that if you like, sir.

Q. (By Mr. Edmonds) You can refer back.

"It is an object of the present invention to obtain these potentially valuable oxidized bodies from ethylene and other olefinic gases by a direct limited oxidation under controlled conditions. Ethylene which is typical of the other olefinic gases is a readily oxidizable substance, but, being endothermically constituted, oxidation is attended with the liberation of much energy and is apt to be too far going, while the direct products of its limited oxidation, ethylene oxid, glycol and chlorhydrin are themselves also readily oxidizable," indicating that we are dealing with things which go up in temperature unless we check them.

Q. Now, the Court asked you if other conditions were set forth there as fully as in Lefort. Are there any other conditions set forth in Lefort? A. No, sir.

The Court: Let me see, to recapitulate. I understand you to say that you think these two patents do directly anticipate; but you do not think any of the other references are an actual anticipation, because they suggest the process, but do not really disclose it.

The Witness: The other references as far as I have indicated anticipate the interaction, the limited oxidation of ethylene and oxygen in presence of silver catalysts, and I call attention to those to indicate that they suggest the question, Why, what did Lefort teach in his patent that these gentlemen had not already done? And if the answer is that he did not give them sufficient directions over and above what they had done, there was an incomplete disclosure. Or, alternatively, you might say, taking Reyerson's experiments, who passed ethylene and oxygen over a silver catalyst, and got carbon dioxide and water, what should he have done, following his process, to get ethylene oxide? If the suggestion is that by taking another silver catalyst which was known, for example, that of Benton & Elgin, then there is a lack of an act of invention on the part of Lefort.

Q. (By Mr. Edmonds) Let me see if we can put that same thought a little differently. Take any one of these prior art disclosures of the catalytic oxidation of ethylene, producing some substance other than ethylene oxide, as, for instance, formaldehyde. Now, if you were to ask yourself, or if anybody were to ask you what change

should be made in the procedure outlined there, so that the resulting product would be ethylene oxide instead of formaldehyde, would it be possible to find the answer to that question in the Lefort patent? A. I don't think so, sir.

Q. Take the Reyerson paper; he performed an oxidation of ethylene and air in the presence of a silver catalyst at an elevated temperature, did he not? A. Yes.

Q. And what did you understand were the products that he got? A. He got carbon dioxide and water.

Q. Now, if anybody had asked you, or if you had asked yourself what to do differently from what is disclosed in that paper as having been done by Reyerson, so that the resulting product instead of being CO_2 and H_2O would have been ethylene oxide, would you have found the answer to that question in the Lefort patent? A. I don't think so, sir.

Mr. Watson: The question is indefinite as to date. I have no idea what date the witness is speaking of.

Mr. Edmonds: Well, will you answer the question with respect to any date from the beginning of the world to 1940?

A. I will answer that in exactly the same way.

Cross Examination.

By Mr. Watson:

Q. You referred in your direct examination to the article by Wurtz, do you recall? A. Yes.

Q. That states, does it not: "Ethylene oxide can not be formed directly by the action of oxygen on ethylene?" A. May I see that?

The Court: Oh, I don't know that you need spend very much time on these prior art articles, because I understand the witness admits— A. That is right, on page 287.

Q. And it also says, "I have in vain endeavored to bring about direct combination of oxygen and ethylene." A. Yes.

Mr. Watson: I just wanted to dwell on a few of those to indicate immateriality.

Q. Do you agree with the statement that before 1931 the published art did not suffice as instruction how to practice direct oxidation of ethylene to produce ethylene oxide? A. I think that one has to bring the McElroy patents into the picture in order to answer the question thoroughly, but I think that there is a very good ground for believing that if one proceeded on the basis of the McElroy patents it would be possible to make ethylene oxide.

Q. That is roundabout, I would like you to answer the question: Do you agree or disagree with the statement that before 1931 the published art did not suffice as an instruction on how to practice direct oxidation of ethylene to produce ethylene oxide? A. I should probably disagree on that basis, on the basis of the McElroy patents * * * [1196-1220].

* * * The Court: Well, I don't know that I understand you. The witness is explaining, as I understand, that in spite of what appears to be considerable lack of specifications, he thinks that at least the later McElroy patent discloses the practice that is involved in the patent in suit. That is the sum and substance of your testimony?

A. That's right * * * [1220-1221].

* * * By Mr. Watson:

Q. You referred to the British Walter patent? A. Yes.

Q. That deals with the production of formaldehyde? A. Formaldehyde from ethylene and oxygen.

Q. No pretence of producing ethylene oxide at all in that patent? A. It does cover the partial oxidation of the hydrocarbons.

Q. But there isn't even an expression of a desire to produce ethylene oxide in that patent, is there? A. I don't think so.

Q. In any event, there is not a faint suggestion of an operating procedure whereby the ethylene oxide can be produced from ethylene and air, is that true? A.

Might I have an interpretation of "faint suggestion," your Honor? I mean, "faint suggestion" is a faint suggestion. I would like to go over it with a microscope * * * [1224-1225].

* * * Q. Willstatter states, doesn't he, that he found his catalysts utterly useless? A. For the preparation of formaldehyde.

Q. And the Reyerson article—I don't know whether you include that within the anticipation or not——

The Court: No, he states he did not.

Mr. Watson: But he spoke of it as a silver catalyst.

Q. As a matter of fact, that was a silverized silica gel catalyst, was it not? A. Yes.

Q. And so stated by the author? A. Yes.

Q. You spoke of results which might flow from using the Benton and Elgin catalyst in place of that silverized silica gel. Was there any foreshadowing in the art, prior to Lefort, that using the Benton and Elgin catalyst would have produced ethylene oxide by direct combination of ethylene and oxygen of the air? A. Not to my knowledge.

Q. Did you ever try to follow the McElroy disclosures? A. No. You mean, practice them?

Q. Did you ever attempt to duplicate them? A. No.

Q. Do you find any statement in either of the McElroy patents of the use of a catalyst of silver at a temperature within the range of 150 to 400 degrees Centigrade * * * [1225-1227]?

* * * A. The two together?

Q. That range of temperature— A. Plus? No.

Q. —and that catalyst that zone of temperature and catalyst? A. No, I do not find that.

Q. Do you find any suggestion in either of McElroy's patents of the use of silver alone as the catalyst? A. Silver alone as the catalyst? Well, if he passed ethylene and oxygen over a silver oxide catalyst in the

temperature range of 150 to 400, you would convert that silver oxide into silver.

Q. And that is no longer an oxide? A. Well, the question is, what is a metal catalyst? With an oxide layer on the surface, is it metal or is it an oxide • • • [1227-1228]?

• • • Q. Do you find a statement in either of the McElroy patents of the use of silver as a catalyst? A. No.

Q. Is it not true that McElroy's statement about the labile oxides concerns the selection of a metal compound with oxygen from which the oxygen is contemplated to be given up, and which oxygen thereafter is contemplated as the source of a reactant? A. I have never met Mr. McElroy, and consequently I can not answer the question.

Q. Irrespective, however, I have asked you whether you found the combination in McElroy of a catalyst having the catalytic characteristics of silver, or silver, and the temperature of 150 to 400 degrees, and you have told me you do not find that combination, is that right? A. Right.

Q. Do you find in the patent a statement of the desirability of using a temperature range of 150 to 400 degrees, even dissociated from the catalyst silver? A. I find the statement of a predetermined temperature range. I find that it is public information that from 100 degrees Centigrade upward the oxidation of the ethylene is beginning in the presence of catalytic agents, and, therefore, this is the range of temperature where I would expect the predetermination to finish up.

Q. And what maximum? A. Oh, with regard to the maximum I would suggest that 400 is a probable suitable maximum because it is at that temperature that ethylene oxide is quite markedly decomposing.

Q. Well, then, you have taken the burning of ethylene to carbon dioxide and water as fixing your 100 degree temperature, have you not • • • [1228-1230]?

• • • A. Yes, I would say that it has been known for nearly a hundred years, if not more than a hundred

years, that ethylene and oxygen, burned in the presence of catalytic agents to carbon dioxide and water at somewhere around 100 degrees Centigrade.

Q. And that would occur at a thousand degrees Centigrade? A. Yes, sir * * * [1230].

* * * Q. Doctor Taylor, do you find that McElroy states any different temperatures for his liquid phase method than for his gaseous phase? A. Yes, he is dealing with the electrolytic oxidation in the liquid phase process and I have never looked whether there is a temperature there.

Q. The question is whether there is any statement in the McElroy patent of a difference in temperature of the two cases, whether it is the liquid phase or gaseous phase? A. I cannot find any temperature in the case of the electrolytic oxidation.

Q. I am not speaking so much of particular temperatures. The question is whether you find any specification of different temperatures in the two cases? A. I cannot find any specification of temperature in the case of electrolytic oxidation.

Q. Well, then, what is the answer to my question? A. My answer to your question would be that in one case he specified a predetermined temperature range and in the other case he does not.

Q. What is that predetermined temperature range? A. The temperature at which ethylene and oxygen would react in contact with the catalyst in question.

Q. What is the statement in the McElroy patent of what that predetermined temperature range is, in figures, if any? A. There isn't any * * * [1234-1235].

* * * Q. How many intermediate oxidation products of ethylene are there? I am asking from a chemical viewpoint? A. A great many.

Q. Is it your position that the McElroy patents singly or together disclose processes for making all of these possible oxidation intermediates? A. No, it doesn't disclose all of those.

Q. How many does it disclose? A. I see three here.

Q. They are? A. The direct products of its limited oxidation, ethylene oxide, glycol and chlorhydrin. That is at the top of page, one, column two.

Q. And line 24 etc., do you find— A. Glycol, chlorhydrin, glycol esters, glycol ethers, ethylene oxide. They are oxygen containing or oxidized derivatives of ethylene.

Q. Is that all? What does the etc. stand for in line 27? A. The other products which we have not mentioned.

Q. Is it your opinion that the McElroy patents disclose operative processes for making all of those? A. It discloses operative processes for making those that he mentioned and those that he included under the term "etc."

Q. That is all the oxidation products possible, chemically, from ethylene? A. Am I to understand that you wish me in one stage——

Q. What is that? A. Am I to understand that you mean in one stage of operation?

Q. I don't care how many stages. A. Of course, it includes carbon dioxide and water as well.

Q. It includes the whole galaxy, doesn't it? A. Yes, I think Doctor Stevenson calculated the number.

Q. Is it your opinion again that McElroy disclosed operative processes for making that whole galaxy? A. He indicates methods whereby those can be produced from ethylene and oxygen.

Q. Did you ever attempt to duplicate the McElroy disclosure to see whether you got any of those? A. No.

Q. Do you know that Mr. McElroy is a patent attorney in Washington, D. C.? A. No.

Mr. Watson: Will you stipulate that, Mr. Edmonds?

Mr. Edmonds: He is a chemist. I have a copy of his letter paper here that says he is a chemist.

Mr. Watson: He is also a patent attorney, isn't he?

Mr. Edmonds: Oh, I think so.

Mr. Watson: And he is alive?

Mr. Edmonds: He is alive. He is getting along in years and because of his increasing years I was unable to induce him to come over here to testify at the trial. There was nothing I would like better than to have him here but he refused.

By Mr. Watson:

Q. Doctor Taylor, you are the co-author of a book with Mr. Rideal? A. Yes.

Q. The book has been referred to in somebody's testimony in the case? A. Yes.

Q. Do you know whether the witness Reyerson, who has testified, is also the co-author of the Reyerson article to which you referred? A. Yes, sir.

Q. Labile oxides include nitrous vapors, don't they? A. Yes.

Q. And they include non-metal compounds then as well as metal. A. Yes.

Q. Do you know whether, in your opinion, turning to the McElroy patent, in your opinion would you get ethylene oxide by passing ethylene and chlorine and steam together as indicated in line 93 to line 95 of the second column of the first page, would you get ethylene oxide that way? A. I don't know, but I don't think so.

Q. As of 1918, the date of this patent, was it known that ethylene and oxygen in the absence of catalysts would combine to make ethylene oxide directly? A. Not to my knowledge.

Q. You never tried it? A. No

Q. The nitrous vapors referred to line 98 include nitric oxide, nitrogen oxide and nitrous anhydride? A. I would hardly think nitrous vapors would include N_2O_5.

Q. It would include NO? A. NO, yes.

Q. And NO_2? A. And N_2O_3.

Q. And N_2O_3? A. Yes.

The Court: Is that all?

Mr. Watson: I have one more question, if the Court please. I hesitated to refer back to the Walter Patent, but there is a mention in it of red heat.

Q. (By Mr. Watson) Dr. Taylor, in the identification of temperatures by colors, red heat means 600 degrees, or in that neighborhood? A. To which place are you referring?

Q. Page 3 of the British Patent, line 31. But it is a general question. I just want an identification of temperature by the term "red heat." A. Red heat, I would say, it is not definitely known where it is, but I should say somewhere around five or six or seven hundred degrees.

Q. And in lines 45 et seq., Walter mentions the 400 degrees C. temperature only as a quotation from Beilstein's handbook. A. Yes * * * [1235-1241].

* * * Redirect Examination.

By Mr. Edmonds:

Q. If you were asked to practice the process described in the McElroy patent, using as the catalyst that labile oxide, silver oxide, and were also to practice the process of the Lefort Reissue, using as the catalyst that catalyst described there as silver oxide, what would you do? A. I would conduct approximately the same experimental investigation in the two cases.

Q. And what would you do about the temperature? A. I would start in something like Professor Lamb did, at a low temperature, and gradually heat up, and find out if there was a reaction, indicated first of all by the rising temperature, and then by an analysis of the product that was obtained.

Q. You have spoken of a Walter British Patent. I show you German Patent 168291. (Handing patent to witness.) Is that for substantially the same thing? A. Yes, sir. The date identifies it.

Mr. Edmonds: That is all.

I offer in evidence this German Patent No. 168291 just identified by the witness, together with the translation of the specification.

(German Patent No. 168291 marked "Defendant's Exhibit No. 83.")

(Translation of specification thereof marked "Defendant's Exhibit No. 84.")

RECROSS EXAMINATION.

By Mr. Watson:

Q. There is no difference between the British Walter and the German, so far as you know, is that true? A. I gather they roughly cover the same material.

Mr. Watson: That is all.

(Examination of witness concluded.)

Mr. Edmonds: I offer in evidence the copy of the Wurtz article referred to by the witness, Dr. Taylor, as Defendant's Exhibit 85.

(Copy of Wurtz article marked "Defendant's Exhibit No. 85.")

Mr. Edmonds: I offer in evidence a copy of Walter British Patent No. 21941 of 1905, also referred to by Dr. Taylor, as Defendant's Exhibit 86.

(Copy of Walter British Patent No. 21941, marked "Defendant's Exhibit No. 86.")

Mr. Edmonds: I also offer in evidence the Willstatter & Bommer article of 1921, together with the translation of it, as Defendant's Exhibits 87 and 88.

(Willstatter & Bommer article of 1921 marked "Defendant's Exhibit No. 87.")

(Translation of Defendant's Exhibit 87 marked "Defendant's Exhibit No. 88.")

Mr. Edmonds: I also offer in evidence a copy of McElroy Patent No. 1253617, as Defendant's Exhibit 89.

(Copy of McElroy Patent No. 1253617, marked "Defendant's Exhibit No. 89.")

Mr. Edmonds: And also a copy of McElroy Patent No. 1308797, as Defendant's Exhibit No. 90.

(Copy of McElroy Patent No. 1308797, marked "Defendant's Exhibit No. 90.")

Mr. Edmonds: I also offer in evidence a copy of the paper written by Dr. Taylor, to which he referred in his testimony, printed in the Journal of the Franklin Institute for 1922. That will be Defendant's Exhibit 91.

(Copy of paper written by Dr. Taylor, printed in the Journal of the Franklin Institute, for 1922, marked "Defendant's Exhibit No. 91.") * * * [1241-1245].

* * * * * *

TESTIMONY OF RAY M. CARTER

Thereupon——

RAY M. CARTER, a witness of lawful age, produced on behalf of the Defendant, having been first duly sworn according to law, was examined and testified as follows:

Direct Examination.

By Mr. Barrows:

Q. How are you employed, Mr. Carter? A. By the U. S. Industrial Alcohol Company in the research laboratory in Stamford, Connecticut.

Q. How long have you been employed by that company? A. I have been employed by that company since 1917.

Q. What briefly has been your technical training and experience? A. I graduated from the University of Maine with the degree of Bachelor of Science in Chemistry and obtained a master's degree in science in the University of Maryland.

Q. How long have you been employed by the U. S. Industrial Alcohol Company? A. Since 1917, that is twenty-three years.

Q. What briefly has been the nature of your work over that period? A. At the start it was largely analy-

tical work and in charge of analytical work and following that research work on various problems of an organic nature.

Q. When did you first become interested in the production of ethylene oxide and glycol? A. Around about 1934.

Q. And you were then in the research laboratory of the company at Stamford? A. That is correct.

Q. How long after that did you continue to be interested in the production of ethylene oxide and glycol? A. I have been interested in that up to this time.

Q. Sometime during the course of that work did you personally try to use silver as a catalyst in the oxidation of ethylene? A. I did.

Q. And at what time was that? A. December, 1935.

Q. How did you happen to use silver as a catalyst at that time? A. We first used silver at that time because we had just finished in the laboratory a process in which silver had been used as a catalyst in the partial oxidation of isopropyl alcohol.

Q. What kind of silver catalyst was that? A. We used that particular form of silver which had been used in that prior work. It was silver in the form of rivets, small rivets.

Q. That was a satisfactory catalyst in this other process? A. Very satisfactory, yes, sir.

Q. What results did you find when you used that as a catalyst to oxidize ethylene? A. We didn't find very good results with that. The maximum conversion of ethylene to ethylene oxide was about 4-3%.

Q. Under what conditions did you obtain that result? A. We used a glass reaction vessel, pyrex tube, essentially like the one shown here yesterday, filled with silver rivets to a depth of 13 inches, and using a mixture of 5 per cent. ethylene and 95 per cent. air. We obtained in our first test a maximum conversion of 2.3 per cent.

Q. Under what conditions? A. The temperature at that time was 417 to 425 degrees Centigrade.

Q. Will you go ahead and describe the experiment you carried out? A. Following that the percentage of ethylene was changed from 5 per cent. to 90 per cent.—90 per cent. ethylene and 10 per cent. of oxygen, and a temperature of 386 to 445, and the conversion was .2 of 1 per cent., as compared with 2.3 previously obtained.

Thinking that the depth of 13 inches was not sufficient, we changed the reactor to 1 inch rather than 1⅛ inch, and got a depth of 25 inches of silver rivets, and were able to get a maximum conversion of 7 per cent., at a temperature of 458 to 465 degrees, with a mixture of 5 per cent. ethylene and 95 per cent. air.

Q. Now, you have used the term "conversion." What does that mean, Dr. Carter? A. By conversion I mean the percentage of input ethylene which is converted to ethylene oxide in the operation.

Q. Now, how did the results obtained by this test compare with the results that you previously obtained in the oxidation of ethylene? A. The conversion of ethylene to ethylene oxide was much greater with the silver rivets than by any other method which we had investigated up to this time.

Q. And had you, prior to this time, investigated various methods of oxidizing ethylene catalytically? A. We had.

Q. Including the use of silver as a catalyst? A. Silver gauze.

Q. And you say up to that time you had never obtained as high a production of ethylene oxide with the catalyst as you obtained without a catalyst? A. Will you repeat that, please?

Q. Well, I will withdraw the question. Will you state again how these results obtained by this experiment in December, 1935, with silver rivets compared with the results previously obtained in oxidizing ethylene without a catalyst? A. They were much higher, much higher than anything we had obtained in catalytic oxidation prior to this time, prior to the use of the silver rivets.

Q. And I believe you referred to a comparison of the oxidation of ethylene with catalysts and the oxidation

of ethylene without catalysts? A. I think you mentioned it in that last question. That is why I was not sure, and asked you to repeat it.

Q. Well, had you, prior to this experiment in December, 1935, carried out experiments in oxidizing ethylene in air without a catalyst? A. We had.

Q. And also with a catalyst? A. That is correct.

Q. A small number, or large number of catalysts? A. Quite a large number of catalysts.

Q. Including silver? A. Including silver.

Q. And how did your results prior to December, 1935, with catalysts, compare with your results without catalysts, in that oxidation? A. Oh, the results with catalysts prior to the silver rivet run were poorer than the results without catalysts.

Q. How did the results obtained with the silver rivets compare with the results previously obtained without a catalyst? A. They were much higher than the conversion obtained previously without catalysts.

Q. Now, did you subsequently investigate the use of silver rivets further after these experiments you have referred to? A. We did. We took another lot of the silver rivets and etched them with nitric acid.

Q. What was the purpose of that? A. We thought we might thereby increase their surface, and perhaps increase their catalytic effect * * * [192-198].

* * * Q. Mr. Carter, you recall that the original describes the addition of water to the air and ethylene used in the process of making ethylene oxide. A. Yes * * * [204-205].

* * * Q. Well, did you, Mr. Carter, carry out any experiments with the addition of water in that process, in comparison with the experiments when water was not used? A. Yes, we did.

Q. Will you describe what experiments you did? A. We had a silver on pumice catalyst.

Q. When did you carry out that experiment? A. That experiment was carried out the 23rd of April, 1936. It.

was a silver pumice catalyst. 5 per cent. ethylene, 95 per cent. air, was passed over that catalyst, at the rate of two feet an hour, the water passing through a water hydrator at approximately 90 degrees Centigrade, which would have given a steam content of about 40 per cent. to the vapors, at 392 to 407 degrees Centigrade. Only 1.8 per cent. of ethylene oxide was the conversion. The hydrator was then by-passed so that no steam went through with the ethylene and air, and at a temperature of 386 to 410, which is very close to the previous one. And the conversion rose to 6.5 of ethylene oxide, showing about three-fold decrease in ethylene oxide with the steam * * * [205-206].

* * * Q. You found that particular catalyst with steam had the effect of very materially decreasing the catalytic effect? A. It had a deterrent effect, yes.

Q. And where is the experiment recorded in your notebook? A. That is in Notebook 1116, at page 19.

Q. And does that end on that page or extend over to the next page? A. It is pages 19 and 20.

Q. Now, did you try water with any other catalyst besides silver on pumice? A. Yes, we tried the effect of steam with silver, on silica gel catalyst.

Q. What time did you try that? A. 21st of April, 1936.

Q. Will you describe what you did there? A. We were using a silver silica gel catalyst in a pyrex tube and a gas mixture containing five per cent. ethylene, and ninety-five per cent. air was passed through the catalyst tube at the rate of 2 cubic feet an hour at a temperature of 284 to 316, a trace of ethylene oxide was formed.

The gas flow was increased to 10 feet an hour and the temperature immediately rose to 450, indicating complete combustion. The ethylene concentration was then cut back to one per cent. and a conversion of 1½ per cent. ethylene oxide was obtained. The inlet gas was then saturated with water as before, giving about 40 per cent. steam concentration and the conversion of ethylene rose to 3.6, about double.

Q. That is in this case the addition of water more than doubled the production of ethylene oxide as compared

with the result obtained without the addition of water. A. Water was beneficial in this case, and it did just that.

Q. At what pages in your notebook is this experiment described? A. It is the same notebook at pages 15 and 16, 17 and 18.

Q. Beginning on what page? A. 15. 16, 17 and 18.

Q. Does page 13 of this notebook relate to this experiment? A. Yes, it does begin on page 13. That is the description of the catalyst chamber.

Q. Now, do your records show how this particular catalyst was prepared? A. Yes.

Q. And do you have a record of that particular preparation? A. Yes, it is in Notebook 1106, I believe.

Q. At what page? A. Page 21.

Mr. Barrows: I offer in evidence as Defendant's Exhibit 7 photostat of pages 13, 15, 16, 17 and 18, showing the record of the experiments described by the witness.

(Pages 13, 15, 16, 17 and 18 of Dr. Carter's Notebook 1106, marked in evidence, "Defendant's Exhibit 7.")

Mr. Barrows: I offer in evidence as Defendant's Exhibit 8, page 21 of the notebook describing the method of preparing the catalyst.

(Page 21 of Dr. Carter's Notebook, marked "Defendant's Exhibit 8.") * * * [207-209].

* * * Mr. Barrows: I think you prepared a brief summary of the notebook records in the form of a connected story on this experiment?

A. Yes.

Mr. Barrows: I offer in evidence a summary of the experiment as Defendant's Exhibit 9.

(Summary of experiments in connected form, marked "Defendant's Exhibit 9.")

Q. Dr. Carter, in this earlier experiment with pumice of April 23, 1936, do you have a description of how the catalyst used in that experiment was prepared? A. That occurs on page 17 of the Notebook 1106.

Mr. Barrows: I offer in evidence photostat copy of pages 19 and 20, describing this experiment of April 23, 1936, as Defendant's Exhibit 10.

(Pages 19 and 20 describing experiment of April 23, 1936, marked "Defendant's Exhibit 10.")

Mr. Barrows: I offer in evidence page 17 of the other notebook describing the preparation of the catalyst as Defendant's Exhibit 11.

(Page 17 of Notebook describing preparation of catalyst used in the experiment of April 23, 1936, marked "Defendant's Exhibit 11.")

Q. And have you similarly prepared by request a summary of this experiment, Dr. Carter? A. Yes, sir, I have.

Mr. Barrows: I offer this summary as Defendant's Exhibit 12.

(Summary of experiment of April 23, 1936, marked "Defendant's Exhibit 12.")

Q. Dr. Carter, prior to the time you carried out these experiments where you added water and operated with and without water, how many carriers had you tried as carriers or supports for silver catalysts? A. We tried only one: pumice.

Q. And the silica gel was the next one you tried? A. Silica gel was the next one.

Q. And what was the next one you tried? A. I believe the next we used was activated alumina.

Q. Did you find that a satisfactory support? A. It was not. It was a very unsatisfactory support.

Q. Did you produce any ethylene oxide whatever with that support? A. It produced none at all.

Q. Did you investigate to see whether you could do anything to make that support satisfactory? A. Yes, we treated and washed the activated alumina with acid in an attempt to remove any alkaline materials that might be there, to see if such a treatment would result in a better catalyst, and it did not.

Q. What was the next carrier support that you tried? A. Infusorial earth.

Q. Did you find that a satisfactory support? A. No, sir, that was not a satisfactory support.

Q. Can you identify the notebook where you tried these supports, alumina and the infusorial earth? A. Yes, sir, alumina is given, starting at page 21, 22, of Book 1116, at pages 27 and 28 of the same notebook.

Q. What was the date of those experiments? A. April 25, 1936, and May 4, 1936.

Q. And have you prepared at my request a summary of these experiments? A. I have.

Q. And can you identify the notebook pages showing how the catalysts were prepared that were used in these tests? A. Yes, sir, in Notebook 1106, page 29, for the first alumina experiment mentioned, and page 33 for the second.

Mr. Barrows: I offer in evidence photostat copies of pages 21 and 22 for the experiment dated April 25, 1936, as Defendant's Exhibit 13.

(Pages of photostat copy, pages 21 and 22, for experiment dated April 25, 1936, marked "Defendant's Exhibit 13.")

Mr. Barrows: I offer in evidence page 29 of the notebook describing the catalyst.

(Page 29 of Notebook describing the catalyst, marked "Defendant's Exhibit 14.")

Mr. Barrows: And a brief description of that experiment is offered in evidence.

(Brief description of experiment of Dr. Carter, April 25, 1936, and May 4, 1936, marked in evidence, "Defendant's Exhibit 15.")

Mr. Barrows: And I offer in evidence pages 27 and 28 of experiment dated May 4, 1936.

(Pages 27 and 28 of the experiment dated May 4, 1936, marked in evidence, "Defendant's Exhibit 16.")

Mr. Barrows: Also page 33, describing the production of the catalyst used in this experiment.

(Page 33, describing preparation of the catalyst used in experiment of May 4, 1936, marked "Defendant's Exhibit 17.")

Mr. Barrows: And the summary sheet describing the experiment.

(Summary sheet describing experiment of May 4, 1936, marked "Defendant's Exhibit 18.")

Mr. Barrows: I also offer in evidence pages 44 and 45, describing experiment of May 4, 1936.

(Pages 44 and 45, marked "Defendant's Exhibit 19.")

Mr. Barrows: And a summary of that experiment.

(Summary of experiment of May 4, 1936, marked "Defendant's Exhibit 20.") * * * [210-2[illegible]].

* * * I offer the pages of the experiment of May 7, 1936, using silver on infusorial earth, comprising pages 29, 30, 31, 33, 34 and 35.

(Pages of experiment of May 7th, 1936, marked "Defendant's Exhibit 21.")

Mr. Barrows: We now offer the page showing the method of preparing the catalyst, page 37 of Mr. Carter's notebook.

(Page 37 of notebook marked "Defendant's Exhibit No. 22.")

Mr. Barrows: And the next a summary of this experiment.

(Summary of experiment, pages 29 to 35 of book 1116, marked "Defendant's Exhibit 23.")

By Mr. Barrows:

Q. Before getting into this detail, can you enumerate other carriers that you tried as support for the silver catalyst and which you found a non-satisfactory support for that purpose? A. Yes, on June 15, 1936, I found that aluminum silicate, silver on that gave no ethylene oxide. Ethylene was burned but no ethylene oxide was formed.

On the 22nd of June we used silver mounted on fire clay and we found a maximum conversion of nine-tenths of a per cent. of ethylene oxide. On the 25th of June we tried silver mounted on glass wool and up to 347 degrees Centigrade there was no reaction. At 453 degrees we had two and a half per cent. conversion to ethylene oxide. On July 3, 1936, we tried silver on asbestos and the maximum conversion which we could get with that was 4.3% at 350 degrees Centigrade. On the 20th of August of 1936 we tried silver on carborundum and our maximum conversion there was 1.4% at 350 degrees. Then on June 11th we prepared silver from tartrate by decomposition by heat, leaving a very light and spongy silver and the maximum conversion with that was 3%. No carrier, just plain spongy silver. We followed that by having silver prepared on pumice and then decomposing that to give us the silver from the tartrate on the pumice and up to 450 degrees Centigrade there was zero conversion of ethylene, starting at a temperature of 200 to 400. Then on the 16th of June, 1936, we took the spongy silver prepared by decomposition of tartrate and laid down on it silver from silver nitrate by evaporation of silver nitrate upon this spongy silver and reduced that with hydrogen and got a maximum conversion of one per cent. of the input ethylene at 350 degrees.

Q. Some of these catalysts you described were silver without a support? A. Yes, the silver with the tartrate was without a support.

Q. And you found no ethylene oxide whatever with some of these experiments? A. That's right.

Q. And the other catalysts were made with silver on various supports? A. Yes.

Q. And many of those gave no ethylene oxide whatever? A. Yes.

Q. And some gave only traces? A. Some gave very small amounts and some gave amounts essentially traces and some gave none at all.

Q. After you had carried out these experiments in which you added water and tried pumice and silica gel, alumina and infusorial earth, did you have occasion to

record your views on the catalysts in your notebook at that time? A. Yes.

Q. Can you identify the pages where you did that? A. Yes, sir, starting on page 53.

Q. What date was that? A. The 28th of May, 1936.

Q. How long had you been working on silver catalysts up to that time? A. I had worked from the latter part of December, 1935, up to this date on the silver catalyst.

Q. How many different catalysts had you made during this time? A. The preceding one was the 49th—no, I beg your pardon.

Q. The 49th experiment? A. The 49th experiment, the 24th catalyst.

Mr. Barrows: I offer in evidence the notes of the witness as of May 28th, 1936 as Defendant's Exhibit 24.

(Notes of Mr. Carter of May 28th, 1936, marked "Defendant's Exhibit No. 24") * * * [218-222].

* * * By the Court:

Q. In that summary do you state what you find to be a satisfactory silver catalyst? A. At that time I had not found a satisfactory silver catalyst. That is the purport of that statement.

By Mr. Barrows:

Q. What were your conclusions as to the effect of the carrier on these catalysts or the method of preparation of the catalysts up to this time? A. Our results had shown that the effect of the carrier was very marked. Obviously there were certain carriers which did not give with silver the catalyst which we expected in the formation of ethylene oxide. Pumice was the best one at the moment. Most of the others were no good at all or so bad as to be of no commercial significance.

Q. What conclusion did you reach as to pumice? Would you say it was the best thing you tried up to that time? A. Pumice catalysts were the best we had found at that time. They had the disadvantage that their activity

decreased very rapidly with use, so rapidly that they were entirely out of the picture as far as commercial operation was concerned at that time. They just would not hold their activities for a long enough period to be of any benefit.

Q. What is pumice chemically? A. It is essentially an aluminum silicate, I believe, but has a lot of other material present in small amounts. I don't recollect right offhand the exact composition of it.

Q. You suggest then that these impurities may have affected the catalyst? A. I thought some of these impurities there may be acting in such a way as to make pumice definitely better than these other materials which we tried and which we had every reason to believe might be produced.

Q. Did you try aluminum silicate in a pure state without the presence of pumice? A. Yes, that's what we did. It represented a pure form of constituent parts of pumice. We thought if we tried it in pure form, we might get some information which would be of help.

Q. In the course of your experiment, Mr. Carter, did you try a copper catalyst as a catalyst for oxidizing ethylene? A. Yes.

Q. What result did you obtain? A. We had complete oxidation of the ethylene to CO_2, no ethylene oxide.

Q. Now, you refer to these experiments with silver gauze. Will you identify the notebook that records those experiments? A. I believe it is No. 1047.

Q. What page in that notebook. A. The silver gauze?

Q. Yes. A. On page 140 and 141.

Q. Do those pages need any preceding pages to explain them? A. Yes, the apparatus is pictured on an earlier page, page 128.

Q. What page should be included to get the whole story of this silver gauze experiment? A. 128 would be needed.

Q. Beginning at page 128 up to the pages you have referred to? A. Yes, pages 140 and 141.

Q. And what results were obtained with this silver gauze experiment? A. The maximum conversion of ethylene to ethylene oxide in this silver gauze experiment was .7 of 1 per cent.

Q. At what temperature? A. At a temperature of 530 to 545.

The Court: What is silver gauze?

The Witness: It is just like a screen, a window screen, only made of silver wire. It was a fine silver wire, made in the shape of a net, network, just like a screen.

Q. (By Mr. Barrows) And is that a common form of silver for catalytic purposes? A. So far as I know, yes, sir.

Q. Now, you say you got about .7 of 1 per cent at a temperature of around 530 to 545? A. Yes, sir.

Q. Is that Centigrade? A. That is Centigrade.

Q. Now, within the range of the Lefort Patent of 150 to 400 degrees, what results were obtained? A. About .05 of a per cent.

Q. .05 of 1 per cent? A. .05 of 1 per cent conversion.

Q. Now, the prior pages that you said were necessary to understand this experiment cover the use of a platinum gauze, and empty, without any gauze at all in it? A. That is correct.

Q. And how did the results obtained with the silver gauze compare with the results obtained without any gauze at all in the apparatus? A. Without any gauze at all we were able to get, with the same apparatus, a conversion of ethylene of about .8 of a per cent, compared to the .7 obtained with the silver gauze.

Q. And at a somewhat higher temperature? A. That was at a temperature of about 610 degrees.

Q. And have you prepared, at my request, a brief summary of this silver gauze experiment? A. Yes, here it is.

Mr. Barrows: I offer in evidence a photostat copy of this, identified by the witness, relating to this silver gauze experiment, and also the witness's summary of this experiment.

(Photostat copy of silver gauze experiment, and witness's summary of same, marked respectively, "Defendant's Exhibit No. 25" and "Defendant Exhibit No. 26.")

Q. (By Mr. Barrows) Mr. Carter, in referring to the silver which you said you tried in your experiments, you said you did get some results there at a rather high temperature? A. Yes.

Q. Considerably higher than the temperature range of the patent in suit, for your best result? A. That is true. The best results were obtained at temperatures above 500 degrees.

Q. And then you tried silver rivets, treated with nitric acid, and got no results at all? A. That is correct.

Q. How did you explain the difference between those results? One gave no ethylene oxide, and one gave some at a high temperature? A. Well, the only things that I know of that have a bearing on that are the fact that the silver rivets which were first used, that is the untreated ones, not nitric acid treated, those rivets had been used previously in this oxidation of isopropyl alcohol; and their surface had the mat effect, and was an oxidized silver surface. That probably indicated a more active silver surface than the original rivets had. Why the nitric acid treated ones should have been inactive, I do not know. They just were.

Mr. Barrows: I find I did not offer in evidence the pages, the notebook records of experiments with silver rivets. I offer the pages from notebook 1074, describing the experiments, as Defendant's Exhibit 27.

(Pages describing experiments, from Notebook 1074, marked "Defendant's Exhibit No. 27.")

The Court: Do I understand you to say that you have or have not found, up to the present time, a satisfactory catalyst?

The Witness: You mean at the present time, right now?

The Court: Yes, now.

The Witness: Oh, yes, we have now.

The Court: Of silver?

The Witness: Yes, we have a satisfactory catalyst as of this date.

Q. (By Mr. Barrows) And how many months did it take you before you arrived at a satisfactory catalyst? A. We arrived at that satisfactory catalyst late in '36, somewhere around about November, 1936, I believe. We started this work in December of 1935, active work on the silver catalyst.

Q. And how many catalysts had you made between the time you started in December, 1935, and the time you found a satisfactory catalyst in 1936? A. I do not remember the exact number, but it was a lot, somewhere around a hundred.

The Court: Well, do I understand it is this catalyst consisting of silver disposed on a carrier of aluminum oxide, bonded by a fusion process?

The Witness: That is correct.

The Court: That is the one.

The Witness: That is correct.

Q. (By Mr. Barrows) And how many other carriers had you tried before you arrived at this one as a satisfactory carrier? A. We had tried probably twice as many as I have related this morning. I do not have the exact number in my mind. At least twice as many than those——

The Court: Twice as many. How many does that come to?

The Witness: Twice as many as we have gone over this morning.

The Court: How many does that come to in number?

The Witness: Now, let me get this clear. Do you want to limit this to what date, the date as of the alundum catalyst, or the date that we have been talking about here?

The Court: No, I was not limiting it to any date.

The Witness: Oh!

The Court: How many experiments, or how many different catalysts did you use before you reached the good one?

The Witness: Approximately one hundred catalysts were made before we reached a good one.

Mr. Fairbank: How many carriers?

Q. (By Mr. Barrows) How many different carriers? A. I would have to estimate that without going through the notebook and look at it.

Q. Could you give a rough estimate? A. I should say probably 40.

The Court: Forty?

The Witness: Yes, sir.

Q. (By Br. Barrows) Now, in addition to the carriers, did you investigate different methods of preparing the catalyst, different chemicals to be used in preparing it, different temperatures of reducing it, or otherwise preparing it, and different percentages of silver on the carriers? A. We did.

Q. What did you find as to the effect of different conditions in preparing the catalyst upon the results obtained by the catalyst? A. We found that the activity of the catalyst varied largely with the way in which the silver was put on it, whether it was put on by evaporation of silver nitrate, or whether it was prepared by deposition of silver hydroxide.

The Court: Now, are those differences included in these one hundred different catalysts you are talking about?

The Witness: Yes, your Honor.

The Court: In other words, they would all be part of these one hundred different catalysts?

The Witness: Yes, your Honor.

The Court: I do not think you need to go all over that again.

Q. (By Mr. Barrows) Well, in addition, Mr. Carter, isn't it also a fact that with an individual catalyst you made many changes in trying to reactivate it, or increase its activity, and subsequently treat it to make it into a catalyst? A. We did * * * [222-235].

* * * Q. (By Mr. Barrows) From your experience, Mr. Carter, in making a silver catalyst for the oxidation of ethylene oxide, what would you say as to the sufficiency of the Lefort Reissue Patent as to how to prepare a satisfactory catalyst? A. In my estimation it is totally insufficient in directions of how to prepare a satisfactory catalyst, silver catalyst.

The Court: You mean it is insufficient both as to describing the way to use the silver, and also the preparation of the supporting carrier?

The Witness: Yes, it neither tells you what support, what to use, how much silver to put on it, how to put that silver on it. It gives you no directions as to the preparation of the support of the catalyst.

The Court: Well, does your answer apply equally well to both patents?

The Witness: In that respect, yes, sir.

The Court: Well, we are only talking about the catalyst.

The Witness: Yes, the supported catalyst.

Q. (By Mr. Barrows) Let me ask you, Mr. Carter, at the time, or when did you first learn about the original Lefort Patent? A. The U. S. original?

Q. Yes. Do you recall it? A. I have no record of the exact date when I first saw it; but it was shortly after the office received a copy of it.

Q. And you recall that there were earlier foreign patents, with a similar disclosure? A. Yes, I have seen those.

Q. Do you recall when you first saw those? A. About 1933.

Q. And did you find anything in those foreign patents, or in the patents in suit, the patent originally granted, to

help you in your experimental work in making ethylene oxide? A. No, sir, we did not.

Mr. Barrows: To complete the offer of the records on the experiment with the silver rivets, I offer page 27, describing the treatment of the rivets, under date of April 21, 1936.

(Page 27, under date of April 21, 1936, describing treatment of silver rivets, marked "Defendant's Exhibit No. 28.")

Mr. Barrows: And pages 43 to 45, describing the later experiment in which those treated rivets were used.

(Pages 43 to 45, describing later experiment in which treated rivets were used, marked "Defendant's Exhibit No. 29.")

Mr. Barrows: And also the summary of the experiments as prepared by the witness.

(Summary of experiments as prepared by witness, marked "Defendant's Exhibit No. 30.")

Q. (By Mr. Barrows) Mr. Carter, referring to the experiments on pumice which had been made, experiments which had been made on pumice as a carrier for the silver, up to the time of your notebook entry at the end of May, 1936, have you prepared brief statements of the different experiments in which you used pumice as a carrier? A. I have.

Q. And in these are identified the pages of the notebook describing those experiments? A. That is correct.

Q. And you have had photostatic copies made of those pages pertinent to these experiments? A. Yes, sir.

Q. And also identify the pages of the notebook describing the catalyst? A. Yes, sir.

Mr. Barrows: I offer in evidence the group of summary reports of the experiments.

(Group of summary reports of experiments, marked "Defendant's Exhibit No. 31.")

Mr. Barrows: And also as one exhibit, the various notebook pages describing these various experiments.

(Various notebook pages describing various experiments, marked "Defendant's Exhibit No. 32.")

Mr. Barrows: And as another exhibit, pages of the notebook describing the preparation.

(Pages of notebook describing preparation, marked "Defendant's Exhibit No. 33.")

Q. (By Mr. Barrows) Mr. Carter, these notebooks that you have produced and discussed were notebooks kept in the regular course of business in recording laboratory experiments as they were made at the time? A. Yes, sir, they are.

Q. And these have been produced from the files of the company and are the original records of the company? A. They are.

Q. Mr. Carter, you have referred to various catalysts, some silver without a carrier and some silver on a carrier, subsequent to the memorandum in your notebook at the end of May, 1936. Have you prepared summary statements of these various experiments? A. I have.

Q. Identifying the notebooks describing them? A. I have.

Mr. Barrows: I offer in evidence these summaries and notebook records as follows—let me ask you, Mr. Carter, these summaries have been made recently?

A. Yes, sir.

Q. And to simplify describing these experiments which are prepared in laboratory form, and not so easy to understand unless you are familiar with the shorthand used in the experiment? A. Yes, it would be a little easier to get the purport of the experiment by reading the summary and not having to go through all the figures in the notebook.

Mr. Barrows: Yes. I offer in evidence the summary of the experiment of June 15, 1936, silver on aluminum silicate, together with the accompanying notebook pages as one exhibit.

(Summary of experiment of June 15, 1936, silver on aluminum silicate, together with notebook pages, marked "Defendant's Exhibit 34.")

Mr. Barrows: Similarly I offer as one exhibit summary of the experiment of June 22, 1936, and accompanying notebook pages dealing with silver on fire clay.

(Summary of experiment of June 22, 1936, and notebook pages, silver on fire clay, marked "Defendant's Exhibit 35.")

Mr. Barrows: I also offer as one exhibit a brief description of the experiment of June 25, 1936, with the accompanying notebook pages dealing with silver on glass wool.

(Brief description of experiment of June 25, 1936, with notebook pages, dealing with silver on glass wool, marked "Defendant's Exhibit 36.")

Mr. Barrows: I offer as one exhibit the summary of experiments of July 3, 1936, July 8, 1936 and July 10, 1936, with accompanying notebook pages, dealing with silver on asbestos.

(Summary of experiments of July 3, 1936, July 8, 1936, July 10, 1936, and notebook pages, dealing with silver on asbestos, marked "Defendant's Exhibit 37.")

Mr. Barrows: I offer as one exhibit the summary and the accompanying notebook pages covering the experiment of August 20, 1936, dealing with silver on carborundum.

(Summary of experiment of August 20, 1936, and notebook pages dealing with silver on carborundum, marked "Defendant's Exhibit 38.")

Mr. Barrows: I offer as one exhibit a summary and the accompanying notebook pages of experiment of June 11, 1936, dealing with spongy silver.

(Summary of experiment of June 11, 1936, and notebook pages dealing with spongy silver, marked "Defendant's Exhibit 39.")

Mr. Barrows: I also offer as one exhibit the summary and the accompanying notebook pages of experiment dated June 16, 1936, dealing with spongy silver impregnated with silver from silver nitrate.

(Summary of experiment of June 16, 1936, and notebook pages, dealing with spongy silver impregnated with silver from silver nitrate, marked "Defendant's Exhibit 40.")

Mr. Barrows: I also offer as one exhibit, summaries of experiments of June 12, July 16 and July 24, 1936, and the accompanying notebook pages, dealing with silver from a decomposition of tartrate used on pumice.

(Summaries of experiments of June 12, July 16 and July 24, 1936, and accompanying notebook pages, dealing with silver from a decomposition of tartrate on pumice, marked "Defendant's Exhibit 41.")

The Court: Mr. Carter, let me see if I understand the sum and substance of your testimony. Is it this, that you were called upon by your company to engage someone skilled in the art, that is, someone that you thought was sufficiently technically trained in the art, to make catalysts in accordance with either one or both of the patents in suit; you were directed by your superiors to engage so many people to do this; do I understand you to say that you feel that the description in the specifications with respect to catalysts and carriers in either one or both of the patents are so incomplete, so insufficient, that upon being given such direction you would feel that you would have to give to those subordinates of yours, these people you were employing for this purpose, a great deal more information or a great many more directions than are contained in either one of the patents before you would feel that they, even though thoroughly skilled in the art, could make a satisfactory silver catalyst?

A. Yes, your Honor. As the result of our experiments I would say that that exactly sums up the situation.

Mr. Barrows: Now, let me follow that up a little bit further, Mr. Carter:

Q. In all these experiments that you have described with silver on various carriers, you have formed mixtures of air and ethylene passing through a chamber containing these catalysts at a temperature within this temperature range of 150 to 400 degrees Centigrade, and

have tested the product from that process to see whether there was any ethylene oxide produced? Is that correct? A. That is true. That is correct.

Q. And in many of these experiments where you have used a silver catalyst in accordance with all these conditions you have produced no ethylene oxide whatever? A. That is correct.

The Court: Well, I meant, of course, in putting my question, that these people were being employed to make these catalysts for this purpose and to do it for a successful purpose.

The Witness: Yes, that's what I understood you to mean.

By Mr. Barrows:

Q. Well, Mr. Carter, isn't it a fact that you had a staff of other men besides yourself working on this very problem over the very period you are referring to? A. Yes, we had several men working on it.

Q. And doing your utmost to get a method of producing ethylene oxide? A. We were certainly trying hard.

Q. And the whole purpose of trying these experiments was to find a catalyst that would work? A. That is correct.

Q. And were there frequent conferences among the men of your laboratory as to what to do to get the final results? * * * [235-246].

* * * A. Yes, we would normally hold conferences on the problems on which we were doing work.

Q. Now, of all these silver catalysts which you have made and tried, do you know of any way of telling what silver catalysts favor the direct oxidation of ethylene to ethylene oxide in a mixture of ethylene and air, and which do not favor the existence of substantial amounts of aldehydes in the products? A. No, I do not. Our experience in the manufacture or testing, rather, of catalysts, led us to believe there was no way that you could tell what catalysts or what preparation of catalysts would be active for this particular reaction until you made it and tried it.

The Court: Now, I understand, again, summarizing your testimony, that you flatly disagree then with Dr. Stevenson, who has said that this process, which is described by this reissue patent, of which the model has been set up and you have seen him analyze the model and the process in the model, he says that anyone skilled in the art, following the teachings of the patent, could work out a satisfactory catalyst and could make the given product in a way that would make it commercially satisfactory. You disagree with that?

A. I think it could be worked out, your Honor. I agree with him to that extent.

The Court: Well, you say it could be worked out only after fretting around?

A. That is correct.

The Court: That one who has not made the experiment that you have already made, that one would have to fret around and try an infinite number of catalysts on an infinite number of carriers?

A. Yes, your Honor, you would have to experiment, largely by the method of cut and try, until you could arrive at what compound gave you good, substantial commercial results.

By Mr. Barrows:

Q. And isn't it a fact also, Mr. Carter, that taking the description of the Lefort patent and the conditions which it tells you to observe and the procedural features of the claim except those that define the catalyst in terms of results, and the conditions in terms of result, that in many of these experiments that you carried out you observed all of those conditions, except as to the functional definition of a catalyst? A. Yes.

Q. That is, you used the silver catalyst under conditions precisely those described in the patent except so far as defined in terms of result? A. That is correct * * * [246-248].

CROSS EXAMINATION.

By Mr. Watson:

Q. By whom were you employed from 1934 to 1937, Dr. Carter? A. By the U. S. Industrial Alcohol Company.

Q. During that interval you worked at the Stamford Laboratories? A. 1933? Yes, I did, from '32 on.

Q. '34 to '37? A. Yes, sir.

Q. And what company owns the Stamford Laboratories? A. That is something beyond by knowledge.

Q. Briefly, what was the nature of your work over the various periods since 1917 up until the time you began the work to which you testified? A. In the first few years after 1917 I did largely analytical work in the research laboratory of the Alcohol Company of Baltimore. Then followed a period of some five or six years during which I was in charge of the Analytical Division of that laboratory. Following that I worked on various research problems that were assigned to me.

Q. Briefly, by the fall of 1935 what experience did you have in the art of catalytic analysis? A. Relatively little.

Q. When did you first study the original U. S. Lefort patent? A. The exact date I don't remember, but it was shortly after it came out.

Q. Dr. Carter, the main product of the U. S. Industrial Alcohol Company has been ethyl alcohol? A. That has been one of its main products.

Q. And that applies for twenty-five years? A. Yes, about.

Q. About how many years has ethyl alcohol been used as the source of ethylene? A. Commercial source of ethylene? I should say from memory about twenty years or more.

Q. Over the period from February, 1935, to November, 1936, as to which you have testified, did you personally do or supervise all of the work of which you have given us records? A. Yes, I think that is a correct statement.

Q. In the period which I have referred to, so far as concerns the work to which you have testified, is it not true that you devoted your attention to the use of silver as a catalyst for the production of ethylene oxide? A. Yes.

Q. Over that same period, however, were you concurrently conducting experiments on other materials or other processes for the production of ethylene oxide? A. Will you please repeat that first date?

Mr. Watson: February, 1935, to November, 1936.

A. Yes, yes, during that period we investigated other processes.

Q. In the fall of 1935 were you informed that the Distillers Company, Ltd., of England was operating this Lefort process with silver? A. No, sir, I was not.

Q. When did you get word that the Distillers Company of England was using silver as a catalyst in the process of making ethylene oxide from ethylene and air? A. I have never received word from them that they have used silver.

Q. Were you informed in the fall of 1935 that the Distillers Company owned the British patent rights to the Lefort process? A. Yes, some time late in 1935, I think it was.

Q. Are the records which you have given us of the work you conducted from February, 1935, to November, 1936, all the records of the work you did with silver in this process and, if not, may we consider them as typical? * * * [630-634].

* * * A. It is typical, certainly, of the period over which you have been given these records.

Q. In February, 1935, you made a test using silver gauze, is that correct? A. No, I did not personally make that test. I know of that test.

Q. How many men were under you? A. At that particular time?

Mr. Watson: Yes.

A. No one was working under me at that particular time.

Q. Well, who conducted those tests on the silver gauze? A. Mr. Fogg.

Q. Do you recall there were two of them? A. Two silver tests?

Q. Two gauzes? A. There was one copper and one silver.

Q. One silver, and there were platinum? A. Platinum.

Q. On the first page of Exhibit 25 there is a note that for maximum activity you anneal the platinum gauze for one hour. Was the purpose of that treatment to make the surface of the gauze more catalytically active? A. We did that in accordance with the instructions of the people from whom we purchased the platinum gauze, and that was the reason for so doing.

Q. That is, the reason which I have stated? A. Yes.

Q. The general procedure for the purpose of improving the surface of the gauze to make it more catalytically active by an appropriate pretreatment was well known to you at the time? A. That particular one, I think, was not well known to me at that time.

Q. Well, you broadly stated the desirability and procedure, suitable procedure, of pretreating the silver gauze to render its surface more catalytically active, that is to render it into finely divided form, was commonly known, wasn't it? A. Yes, I knew of the fact of finely divided silver.

Q. Do you recall why you failed to use any such pretreatment with the silver gauze in these experiments although you did use it in the employment of platinum? A. The pretreatment was given the silver gauze.

Q. Is that indicated in your notebook? A. I don't think it is indicated in the notebook.

Q. You mean you think it is not indicated in any of the records which you have given us? A. Most likely it is not. I am not sure of that but I don't think it is.

Q. Were you familiar generally with the article by Bone and Wheeler which gave a description and picture of silver gauze to increase its activity for catalytic use? A. I don't remember whether I was familiar with it at that time or not.

Q. In the test on silver gauze in Exhibit 25, your records give a sketch of the apparatus. Are we to understand that you used one layer of silver gauze across the catalyst tube; in other words, clamped between flanges of the pipe and then merely passed the gas mixture through that layer, through the spot, whose dimensions you have given as about the size of a nickel, a five cent piece? A. I think yes, that is correct.

Q. Do you recall that in this silver gauze experiment you made no test for carbon dioxide in the exit gases while the temperature of the silver gauze was as low as 400 degrees Centigrade? A. I don't recall that.

Q. Will you please turn to that Exhibit 25 and tell me whether as far as that notebook is concerned it is not true that there is no test for carbon dioxide as low as 400 degrees? A. What is the page, please, Mr. Watson?

Q. The last page, I think, of that exhibit. I think only the last page and perhaps the first relate to silver. The balance is platinum? A. No, no indication of carbon dioxide appears there.

Q. So as far as the records indicate, you could not tell whether any reaction was occurring between the ethylene and air at temperatures below 400 degrees? A. There is one gas analysis recorded, there are two gas analyses recorded here, one is above 400 and the other, no temperature is recorded in the preheater, the preheater was cold.

Q. Before you undertook this February 1935 work, did you familiarize yourself with the literature on the subject of known methods of making active silver catalysts? A. I had started to. I was not entirely familiar with it at the time this was done.

Q. Were you fairly familiar with it when you returned to work on this general subject in December, 1935 and February 1936? A. I was more familiar with it.

Q. You were what? A. I was more familiar with it.

Q. Did you at that time find that one commonly regarded method of such a preparation was by taking silver nitrate and precipitating silver oxide and then reducing it with hydrogen? A. Yes.

Q. After your February work, is it not true that as far as your records are concerned, your next test was in December of that year? A. Yes, the next test.

Q. And that was the test on silver rivets? A. That is correct.

Q. What was the size of those rivets? A. I don't remember the exact dimensions. They were about three-sixteenth inches long and the body of the rivet about three-thirty-seconds of an inch. They were quite small.

Q. In Exhibit 27 which concerns your silver rivet experiments, you obtained some ethylene oxide, did you not? A. I did.

Q. And you produced that product even though the temperature through the catalyst bed was by no means uniform, is that true? A. That is correct.

Q. In that experiment also, I think you stated or your notebook records the fact that about two-thirds of your silver rivets had been previously used and had a mat surface. Now, if the other third had had a similar surface, wouldn't the tendency have been to produce more ethylene oxide? A. That, I cannot state for a fact because I never used entirely new and previously unused silver rivets.

Q. As to the later tests reported, are we to assume that all the notes in the notebook were entered on the dates they appear? A. Unless it is stated to the contrary.

Q. Please refer to Exhibit 36. Apparently there is some slight discrepancy there which I wish you would clear on the first page; that is page 77, which is dated June 25, 1936. It refers to the operation and speaks of the catalyst employed, as I read it, E-44, whereas on pages 87 which seems to be with reference to the preparation of that catalyst, it appears that the date was October 17th, 1936, several months later? A. That is hardly possible that that could be a catalyst made in October and

tested in June. I expect that is an error in reporting the facts in the notebook.

Q. I agree with you that there is an error of some kind of course. I wonder whether that may indicate that the catalyst numbers in these reports were entered in the notebook at later dates? A. Not that late. Sometimes we would make one catalyst, it might be made today and entered in the book the next day.

Q. Well, then, you have no explanation for this obvious error? A. No, it might be worth while to look through the catalyst book and see whether the dates would not eliminate that. It was just an error.

Mr. Barrows: Will you look through the catalyst book you refer to and see what page you find that on and what the context is?

The Witness: Well, the two preceding entries, one for E-42 that was made June 15th, 1936, and E-43 was on June 16th, 1936 and E-44 bears the notation October 17, 1936 and the one on the next page is October 18, 1936, both in my handwriting, and then drops back to June 23, 1936 and then goes on to June 24th. I think I must have labored under a misapprehension as to months when I wrote that.

Q. Under whose direction or at whose request were all these tests run? A. Why, in the main at the request of the head of the laboratory.

Q. Who is that? A. Doctor Metzger.

Q. Exhibit 32 mentions they were made at the suggestion of Mr. Balcar. Who is he? A. I believe he bears the title of assistant chief chemist for the Air Reduction Company.

Q. And Mr. Carter, who is Doctor Metzger, so we can have him identified? A. He is in charge of research and development for the U. S. Industrial Alcohol Company and vice president of the Air Reduction Company in charge of research and development for that company.

Q. He has been in the Court room during the trial? A. I believe he has.

Q. By the way, where did you receive your doctor's degree? A. I haven't. I did not so testify. You have been calling me Doctor.

Q. You did not want to take advantage of me? I merely adopted the terminology by which you have been addressed by your own counsel? A. I gave my counsel correction that appeared in the record in that respect. I don't know whether he has given that to you or not.

Q. No, he hasn't. A. That will appear in due course of time, I am sure.

Q. In your direct examination, I understood you to tell the Court despite all of your efforts you had been unable to find a satisfactory catalyst for reacting ethylene and air to produce ethylene oxide before September 1936. Is that the impression you wish to convey to the Court? A. I don't know that those are my exact words but that is certainly the sense of what I would have meant. Up to that time I had not found a successful catalyst for the commercial production of ethylene oxide from ethylene and oxygen.

Q. Well, you mean up to that time you had not found a successful process; that is, a successful catalyst for use in demonstrating the operativeness of the process for producing ethylene oxide without aldehydes by passing the mixture of ethylene and air over the catalyst? A. The commercial operativeness, do you refer to the commercial operativeness? We had not yet discovered a catalyst. We were able before that time to make small amounts of ethylene oxide.

Q. What do you mean by small amounts? A. Oh,——

Q. Are you speaking in pounds or gallons, quarts or percentages? A. I mean amounts which would appear to be too small for commercial operation.

Q. Well, of course, the amounts involved, the amounts produced, I assume, depend a good deal on the size of your apparatus, and how long you conduct the operation; is that true? A. Well, we would judge that largely on the percentage of ethylene converted to ethylene oxide, regardless of the size of the apparatus.

Q. So what you intended to give the Court the impression of was that until November, 1936, you had not been able to find a silver catalyst that would effect the conversion of the ethylene into ethylene oxide at a high enough efficiency or yield per pass. A. High enough to be satisfactory for commercial use, that is correct.

Q. And what figures on conversion and efficiency do you wish us to understand, so that we can understand what you mean by saying they were not commercially satisfactory? A. I don't know that I can give a single figure which would satisfy that requirement, because the commercial success would depend upon some other factors besides the mere conversion. But certainly one ought to have a conversion of, let us say, 30 per cent., or better, I should imagine, in order to envisage commercial use.

Q. And that is single pass yield? A. I understood you to ask about conversion now. By yield, do you mean what I mean by conversion?

Q. Well, define your word "conversion"? A. I defined the word "conversion" the other day by saying it was the percentage of ethylene fed into the apparatus which appeared as ethylene oxide as an exit product.

Q. How would you define the word "efficiency?" A. Well, I have usually in my notebook, I have used the word "yield" as a rule instead of efficiency, meaning the percentage of the ethylene consumed, which has appeared as ethylene oxide.

Q. You call that yield? A. I call that yield.

Q. And what would that percentage be, within your present definition of satisfactory for commercial purposes? A. That would vary somewhat. Probably it would vary between limits of, let us say, 45, 55, 60 per cent.

Q. Well, then, are we now to understand that long before November, 1936, you had obtained silver catalysts satisfactory for conducting this process, when we define the word "satisfactory" by saying that it proved the operativeness of the process? A. You have not sufficiently yet defined the word "satisfactory," I don't think.

Q. Well, I will make it— A. I can not answer that question "Yes" or "No" until that point is clear.

Q. Did you want to give the Court the impression that not until November, 1936, were you able to get a silver catalyst which you used in your laboratory tests, and over which you passed ethylene and air, at a temperature between 150 and 400 degrees, to obtain ethylene oxide in yields up to 40 per cent.? A. Well, the one other point which makes the catalyst operative commercially would be that those yields would be maintained at a reasonable figure.

Q. Well, that does not apply. I have excluded what you have called commercial operations from my question. I am asking you now whether for informative purposes of teaching a skilled chemist how to conduct this process of passing ethylene and air over a silver catalyst, within the temperature range stated, from that viewpoint, had you done nothing before November, 1936, to satisfy you of the operativeness, to demonstrate the operativeness of the process? A. If you define the operativeness of the process as a production of a substantial and economic amount of ethylene oxide, we had not. Defined as those——

Q. All right, your term has been 45 to 55 per cent. yield. Now, before then had you obtained 44 per cent. yield? A. I don't remember whether we had or not. I would have to look up the records to see whether those figures are * * * [634-649].

* * * Q. Will you refer, please, to Exhibit 31? That is a group of summary sheets on your experiments with silver on pumice, is it not? A. It is.

Q. Is it not true that in the experiment of February 18th, the fifth day, at a temperature of 310 to 322 degrees Centigrade you noted you were obtaining a yield of 15.8 per cent.? A. Page 8?

Mr. Watson: Yes.

A. I do not seem to see it. 322 degrees? I just can not seem to find the particular records to which you refer.

(Mr. Watson indicates to the witness.)

The Witness: Oh. 15.8 per cent.? I understood you to say 50. Yes.

Q. That yield was 19.8. Excuse me. Wasn't it? A. 19.8, yes.

Q. And the consumption was 25 per cent.? A. That's correct, that's what is stated.

Q. Can you calculate what the efficiency was from those figures? A. Yes, if you divide 24 into 19.8.

Q. That shows an efficiency of 82½ per cent., doesn't it? A. Approximately.

Q. Well, we will take that subject to your correction later on, if you wish. A. It is approximately correct.

Q. In the same exhibit on the eighth day, apparently beginning March 24, you were obtaining at 322 degrees a yield of 35.9 per cent.? A. March 24 the eighth day appearing in this tabulation? Not 35, no.

Mr. Watson: No, that is the eight day test, but it is the fifth day I am referring to.

Mr. Barrows: What date is that, Mr. Watson?

Mr. Watson: March 24.

A. 35.9.

Q. And that efficiency was about 69 per cent., wasn't it? A. It would be roughly that, yes.

Q. And the sixth day the yield was 29.2 per cent.? A. Conversion 29.2.

Q. Well, that is conversion to ethylene oxide? A. Conversion to ethylene oxide.

Q. And the percentage consumed was 52? A. That's right.

Q. So the efficiency was about 56 per cent.? A. Yes.

Q. And on the next day the conversion was 33.2 per cent.? A. That is correct.

Q. And the per cent. consumed was 52 per cent.? A. That's right.

Q. And the efficiency, therefore, was about 64 per cent.? A. It would be a little less than 64.

Q. Well, 63.9, was it not? A. Yes, about 64.

Q. And on the eighth day the per cent. of conversion was 31.8, per cent. consumed 64, which would give an efficiency of about 50 per cent.? A. Yes.

Q. And the third day you had per cent. conversion of 37.7 and a per cent. consumed of 60, which would give an efficiency of about 63 per cent.? A. That would be right.

Q. Well, now, we can calculate the efficiencies in all these cases by the mathematics you just explained, can't we? A. Yes.

Q. I mean I do not want to go over all of these figures with you, because they are in the record and we can make our own calculations for efficiencies? A. Yes, you can calculate those.

Q. Well, then, it appears in this very first group of tests when you used silver on a support over a period of a week's operation or longer you were getting ethylene oxide yields, conversion per cents. of over 30 per cent., consumption figures over 50 per cent., and efficiencies over 50 per cent., is that right? A. Correct.

The Court: Well, now, why did that not satisfy you? The answer to that question I would be interested in.

The Witness: The answer to that is we could not repeat those with silver on pumice and get these results.

The Court: You mean it was not a question of getting isolated satisfactory results but continuous operation?

The Witness: That is just it exactly. We would make up another catalyst in the same fashion and we would not necessarily get the same results at all.

Q. Well, did your repeated tests prove the point you just made: that you did not get continuous satisfactory results? A. Yes.

The Court: With the same catalyst?

A. Yes.

By Mr. Watson:

Q. Well, do you mean that literally, Mr. Carter? That if you took the same support material, put on the same silver catalyst in the same way, you would not get duplication of results? A. That certainly was true with some of the early work with pumice.

Q. Well, was that because of deliberate variations in your procedure of putting the silver on the pumice? A. No. No, that does not follow.

Q. Was it because of deliberate variations in the amount of silver deposited? A. No, those results would have——

Q. In this particular instance with the pumice don't you explain it and didn't you explain it to yourself at the time as the result of a rather miscellaneous assortment of pumice? A. A miscellaneous what? I beg your pardon?

Q. Assortment of pumice. Didn't you think the variance of results followed from variations in the pumice? A. I don't see how that could possibly have been. We tried various lots of pumice. I do not think the difference arose from the variation in the particular lot.

Q. Let me ask you this general question on this subject, then: As the result of this first week's test of silver on pumice, when you got the yield which you have indicated and the efficiencies which you have indicated in the production of ethylene oxide, weren't you satisfied in your own mind at that moment that it was possible to make a silver catalyst which under this process would directly combine ethylene and molecular oxygen to produce ethylene oxide without substantial aldehyde formation? A. That is true if you do not limit the amount of ethylene oxide formed. We made ethylene oxide. The records show that.

Q. And it is true if you do not limit it in the per cent. beyond the figures recorded in your records? That is also true, isn't it? It seems to me it is inescapably true. A. The figures recorded in the records show that after about eight or nine days' operation the activity expressed by conversion had fallen from a maximum of

37.7 to a minimum of 30.7. That is 7 out of 31, or a 20 per cent. loss.

Q. Now, I will have to have an answer to my question. As of that date you were thoroughly convinced, were you not, that it was possible to make an active surface silver catalyst over which you could pass ethylene and air at a temperature within the range we have spoken of and produce ethylene oxide in substantial percentages with satisfactory efficiencies? Isn't that true? A. That experiment led us to think that it would be possible to do it.

Q. Well, more than that, that experiment led you to believe that you had done it, didn't it? A. No. No.

Q. Did you refuse to credit your own figures? A. These figures were perfectly good as far as the initial activity of the catalyst was concerned, but that 14 per cent. drop in conversion over a period of nine days led us to believe that the thing commercially would be of no success. Therefore, we continued the experiment.

Mr. Watson: Well, I will let the records speak for themselves.

Q. Please turn to Exhibits 21 to 23 dated about the 7th of May, 1936. Here you added sodium hydroxide to form silver oxide, is that right? A. Yes.

Q. And your summary says that after the precipitation by sodium hydroxide the catalyst was washed with an alkali. That is your summary, isn't it? A. That's the way it reads.

Q. The notebook statement indicates the contrary, doesn't it? A. Whereabouts?

Q. Page 37 of Exhibit 22 shows that the mass was washed with water only until no alkali could be tasted, doesn't it? A. I don't seem to have the page——

Q. Page 37 of Exhibit 22? A. I think that is tested.

Q. How can it be that when the notes go on to say that the wash water turned litmus blue, which does show that alkali was still present, doesn't it? A. Yes, and that is followed by the statement here that it had a dark opalescence, which looked like silver that had been reduced by light.

Mr. Watson: I will ask defendant's counsel if he will agree without calling a handwriting expert that the word is tasted.

(Discussion follows at the trial table.)

Q. I hand you the original notebook about which you have previously been examined from a photostatic copy. Is this in your handwriting, page 37? A. That is.

Q. Well, now, don't you think the word on line 8 is tasted? A. I think that is, it is much clearer than on the photostat.

Q. Now, what was your answer on what was proved by the fact that the wash water turned litmus blue? A. I called attention to that but you had not completed the sentence.

Q. That proved, didn't it, that there was still alkali present? A. Yes, that proved that there was alkali present.

Q. It would have been a simple matter to make sure that all of the alkali was removed from the silver oxide by first precipitating the silver oxide from the nitrate, then washing the precipitate thoroughly and then mixing a slurry of the precipitate with infusorial earth, isn't that true? A. It could have been done in that way, yes.

Q. By the way, you saw the demonstration conducted two weeks ago at South Charleston? A. I did.

Q. And you saw how that silver was prepared on the carrier? A. Yes.

Q. That was a simple, normal, obvious procedure, wasn't it? A. Yes, it was very carefully prepared.

Q. And it was a simple, obvious way to do so? A. It was a simple way, yes.

Q. Well, it was an obvious way in that it had been taught for fifteen years or more, isn't that true? A. It has never been taught me for fifteen years to put 32% silver on filtros.

Q. But the procedure followed? A. The procedure of precipitating the silver you referred to?

Q. Yes. A. That is a perfectly normal procedure.

Q. Everything was normal then except the amount of the silver? A. The method of precipitating the silver was perfectly normal.

Q. Was everything else about it perfectly normal and well known in your opinion except the amount of the silver? A. Well, I wasn't familiar with that choice of support.

Q. Suppose we use carborundum. You are familiar with such a support? A. Carborundum?

Q. Yes. A. Yes, I think I have made a catalyst on carborundum.

Q. Did it work? A. My recollection is it did not work well.

Q. Then with the possible exception of the particular inert carrier filtros and the amount of the silver, 32% or whatever it was, you think everything else about it was well known, common, usual procedure of preparing a silver catalyst on a support? A. Yes, I think it probably was.

Q. And your answer would be the same when we put the date say ten years back? A. I am not so certain of that because I don't just recollect just what the state of the art was exactly ten years ago.

Q. Well, you are familiar with the Benton and Elgin disclosure of how to prepare an active surface silver catalyst? A. I have read that, yes.

Q. And that is dated— A. I don't remember the exact date, about 1930 or thereabouts, I think.

Q. Now, in three of your experiments you used silver oxide, didn't you? A. We used silver oxide. I don't remember whether it was exactly three.

Q. Four of them, I believe. Refer to Exhibit 31, the sheet dated March 16th. Here you added silver nitrate solution to nitric acid washed pumice? A. Yes, sir.

Q. And you evaporated it to near dryness and then added sodium hydroxide? A. That is true.

Q. The mixture was washed and your records show the wash still indicated the presence of unreacted silver nitrate, is that correct? A. Yes.

Q. In Exhibit 32, comprising pages 180 to 182 of your notebook, that does not tell us, I believe, how you made your silver oxide, is that right? A. Apparently not.

Q. You used it, however, in such a way that the catalyst got pretty well channeled, is that correct? A. It was of very short depth, I remember that.

Q. And your experimenter noted in his records, did he not, that it was hardly a fair test on that account? A. That is correct.

Q. Nevertheless you got 4.3% conversion of ethylene oxide, didn't you? A. Yes.

Q. Now, refer to Exhibit 34. Here you make silver nitrate with chemically pure aluminum silicate and precipitated the silver oxide with sodium hydroxide wash, is that right? A. That is right.

Q. Was your aluminum silicate fused or fritted into active form? A. Aluminum silicate?

Q. Yes. A. That was bought, I think, from—it was a C. P. product. I know nothing about it other than that it was finely powdered, that is all I know about it.

Q. You don't know whether it had been subjected to high temperature? A. I don't know anything about that.

Q. The pumice you used as supports, however, was in a prefused form? A. The natural form in which it came to us.

Q. What was that form? A. Crushed to about quarter mesh size.

Q. It had not been fused? A. My understanding of pumice is that it is a product of nature which at some time has been fused.

Q. In other words, naturally it has been subjected to a high temperature before it is going to be subjected to use as a support, is that right? A. As far as I know it is.

Q. At that time had you consulted the literature to find out whether activated alumina, for example, destroyed ethylene oxide at the temperatures involved in the use, at 150 to 400 degrees? A. Destroyed ethylene oxide?

Q. Yes. A. No, I did not try specifically for ethylene oxide.

Q. I say at that time had you examined the literature to find out whether that was a fact? A. Yes, activated alumina? No, I never had examined the literature for that.

Q. Did you ever examine the literature to see whether that was a characteristic of silica gel also? A. To decompose ethylene oxide?

Q. Yes. A. No, I don't know anything about that.

Q. Have you received information in any fashion to acquaint you with the fact that such activated alumina or silica gel within the language of activities we have been dealing with was not inert; that is, it did act to destroy, to transform ethylene oxide into acetaldehyde? A. No, I received no such information.

Q. What is your opinion on that? A. I have never tried that particular experiment. I do know that both these are inert as far as the oxidation of ethylene is concerned.

Q. Of course, they would not be inert in this process, would they, if they were active in their own right to destroy the very product which you desire to obtain in the reaction? A. No, if that is the case, I don't know that that is the case.

Q. When you undertook this work, did you consult the literature as to suitable supporting material? A. I don't remember consulting the literature, no. I think I depended largely on my own knowledge.

Q. And your own knowledge included a knowledge of the desirability of using an inert support? A. Inert, as far as the combination of the support and silver was concerned, the support and catalyst.

Q. And it included the knowledge of the obvious desirability of using a support which would have some mechanical strength; is that right? A. That would be necessary, yes.

Q. And that support should have a surface to which the catalyst silver could adhere well; is that right? A. That would be an essential to a good catalyst, yes.

Q. And you knew that it was obviously desirable to use a support so that your finished silver catalyst would be, say, in granular or broken pieces, so that the gas flowing through the catalyst bed would be well mixed, and would not channel its course through the bed. A. Yes.

Q. In June, 1936, your records show that you did some work on silver tartrate. Do you recall that? A. Yes, I do.

Q. Please refer to Exhibit 39, which is your summary sheet dated as of June 11, 1936 (exhibit handed to witness). In this test you attempted to decompose this tartrate to silver by heating it on an old unglazed plate; is that right? A. It was an unglazed plate, I don't remember the old. It may have been.

Q. The note says "old," doesn't it? A. It says "old" here.

Q. As far as the notes indicate, there might have been something on that old plate which might have poisoned the silver as a catalyst? A. Well, the notes don't so indicate.

Q. They don't exclude that possibility either, do they? A. No.

Q. No. Your method of preparing that silver catalyst was simply by putting it on this old unglazed plate, in the open air, over a Bunsen burner. Is that right? A. Correct.

Q. Is it not true that the experimenter's notes indicate that the heat necessary to decompose the tartrate was so strong as to render the catalyst relatively inactive? Page 104. A. He suggested that that might be one cause for its inactivity.

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Q. At that time didn't you know other silver compounds which would decompose under lower temperature than the tartrate? A. Yes, I believe that at that time I tried oxalate. I don't know just what its temperature of decomposition is. But it is probably a little lower than the tartrate.

Q. You have not, however, produced such records among those presented here? A. Well, I was unfortunate in that respect. I could not seem to hold silver where I could use it thereafter.

Q. Is it not true that in your experiments conducted after about the first of May, 1936, your results tended to go down hill? A. In what respect do you mean, Mr. Watson? Do you mean in respect of conversion.

Q. Yields and efficiency? A. I was not aware that it was a matter of time. We may have been using certain catalysts and certain supports which would have resulted in that, but I would have to look them over to see whether they fall in that date or not * * * [656-677].

* * * Q. You spoke in your direct examination of about 40 supports you tried, and I think your records indicate or show about a dozen or so. Do you want to give us a list of the remainder A. That list of 40 went up to the time of the present. I answered his Honor on that matter, and that included, those included some that have been made recently.

Q. I see. So that the only ones you had used up to November, 1936, were those that are already in the record? A. I think there may be some others that are not included in the record, but it would be a relatively small number.

The Court: Well, it is only about twelve in place of forty?

The Witness: Forty are the ones that have been used altogether.

The Court: But up to November?

The Witness: Up to November I do not recall the exact number but——

Mr. Fairbank: I would say about a dozen would be about right.

The Witness: About a dozen would be about right * * * [688-689].

* * * * * *

Thereupon——

RAY M. CARTER, a witness recalled by the defendant, testified further as follows:

DIRECT EXAMINATION.

By Mr. Barrows:

Q. Mr. Carter, during your prior examination, you described experiments made with the silver on pumice as a catalyst and extending over a period from February 1, 1936, to the latter part of May and in your note book record of May 28, 1936, Defendant's Exhibit 24, you summarized your work up to that time and outlined further items of investigation. Did you continue your work on a silver on pumice catalyst after May 28, 1936? A. I did.

Q. How long did you continue your work with the silver on pumice catalyst? A. That work was continued until near the end of November, 1936.

Q. What was the nature of that continued work? A. That work consisted of a continuation of a study of the method of preparation of a silver on pumice catalyst. Studies were made of the method of deposition of the silver, the amount of the silver and the method of reduction of the silver compound. Attempts were made to prolong the life of the catalyst by various means of revivification, and in general to improve the operating conditions with this kind of a catalyst.

Q. During this period from May to November did you try other catalysts than silver on pumice? A. Yes, we tried silver on various other supports than pumice.

Q. With what results? A. Several supports gave catalysts which produced little or no ethylene oxide. None of them appeared to be as good as pumice.

Q. Why were you concerned in trying various supports for the silver? A. The silver on pumice catalyst required a high percentage of silver, around fifty per cent. for best results. We realized that it would not be feasible to use a straight silver catalyst without a support in a commercial process and that it would be necessary to use a supported silver catalyst. We hoped to find a better support than pumice on which a lower percentage of silver could be used and which would have a longer life and a higher capacity for production of ethylene oxide than the silver on pumice catalyst.

Q. How did you control the temperature in your experiments on the silver on pumice catalyst? A. Up to the early part of September we used an electrically heated apparatus and controlled the temperature by regulation of the electric current. We also used an apparatus in some experiments where the reactor was immersed in a fused salt bath. Early in September we installed a small reactor with a jacket surrounding it containing diphenyl oxide. This was installed to give better heat control along the lines which would be used in eventual commercial operation.

Q. What followed the use of the silver on pumice catalyst in November, 1936, and what was the reason for discontinuing the use of such catalyst? A. In November, on checking back over the various carriers or supports that had been tried to see if any of them offered promise for further investigation and as possible improvements on pumice, I found that the best results previously obtained with any other carrier was with alundum back in July and I started to make a further investigation of this material as a catalyst support.

Q. With what result? A. This support for the silver proved to be so much better than pumice that further work with pumice was stopped and we made rapid progress from that time on in our development work.

Q. What advantages did you find the alundum carrier to have and what progress did you make from that point on in your work? A. We found that much less silver was required on the alundum support than on pumice and with ten per cent. of silver on alundum

we obtained radically better results than we ever had obtained with the pumice carrier. We found that the productive capacity of the catalyst was far greater than could be obtained with pumice. A greatly increased flow of gases and a greatly increased production of ethylene oxide could be obtained. We found that the catalyst did not deteriorate with age as did the silver on pumice catalyst and maintained its high activity for a long period of time. With the decreased amount of silver required, the greatly increased production capacity and the long life of the catalyst, we realized at once that we had found what we had been hunting for and had a commercial catalyst. From that point on, progress was rapid.

The apparatus built and used early in September, 1936, and which had a diphenyl oxide jacket for temperature control was one-half inch in diameter and four feet long. Early in December we installed and operated a similar tube sixteen feet long and later in December, 1936, another tube two inches in diameter and sixteen feet long, both being provided with a diphenyl oxide jacket for temperature control. These tubes were designed to obtain operating data for use in designing a larger scale pilot plant. In January, 1937, we installed and operated another converter with a tube one inch in diameter and sixteen feet long. During April, 1937, the production obtained from the two-inch tube unit was at the rate of about fourteen pounds per day with a yield of about 65 per cent ethylene oxide.

In April, 1937, plans were started for a larger pilot plant containing twelve tubes, two inches in diameter by sixteen feet long, and at the same time preliminary plans and estimates were started on a commercial plant, based on the results that had been obtained on the single tube unit.

Q. Did you take steps to protect by patent the production of ethylene oxide with the use of the silver on alundum catalyst? A. Yes. A patent application was filed for the production of ethylene oxide with this catalyst on May 8, 1937, serial number 141422, which matured into United States Patent 2,125,333 on August 2, 1938.

Q. Will you briefly summarize your work on the production of ethylene oxide with the use of a silver catalyst prior to the middle of May, 1937. A. Throughout the year 1936 and up to that date we had carried out an extended investigation of the process, investigating numerous silver catalysts and had succeeded in discovering and developing a satisfactory silver catalyst with alundum as a support or carrier. Except for a few experiments, the process was carried out with the use of a mixture of ethylene and air and without the addition of water to the gaseous mixture passed over the silver catalyst for the production of ethylene oxide therefrom and at temperatures generally between 250 and 350 degrees C. Except for isolated experiments, hydrogen was not admixed with the gases. The commercial catalyst which was developed of silver on alundum as a support and the results obtained with this catalyst were such that rapid development work had taken place and at the time referred to, the middle of May, 1937, plans for a larger pilot plant were under way as well as preliminary plans and estimates for a commercial plant.

Cross Examination.

By Mr. Watson:

Q. In your direct examination it was not your intention to contradict any of the statements of the witness, Adams, was it? A. My direct examination was made before it. I had no notion——

Mr. Watson: This one (indicating statement), this examination right here.

A. Oh, that one? No, sir.

Q. Your statement, so far as there may be discrepancies between this and Mr. Adams' statement, may we not prefer his? A. I do not know that there are any discrepancies there.

Q. I am asking you, however, if there are, may we not take his word? A. You can take whichever word you choose; I have nothing to say about what choice you should make as to that.

Q. In one of these answers you said that you realized it would not be feasible to use a straight silver catalyst. Was that for financial reasons? A. That was for two reasons: we felt that the use of a straight silver, unsupported silver catalyst, would be much more expensive than the supported silver, and, in the second place, we felt there was a possibility that unsupported silver would deteriorate at a faster rate than supported silver.

Q. Did you ever make tests on silver pellets? A. Pure silver?

Q. Yes, up until this time, that is, the date you are referring to, November, 1936? A. Not silver pelleted in the sense, I think, that you refer to. We had used silver made from silver oxide.

Q. And up until November, 1936, your experimental apparatus had been on a size no larger, perhaps not so large, as Plaintiff's Exhibit 3 in the courtroom, is that correct? A. (After looking at Plaintiff's Exhibit 3) It was about the same length but with somewhat smaller diameter tube, in the case of one of the pieces of apparatus; others were essentially of the same diameter.

Q. The patent you referred to suggests the use of silver up to 50 per cent by weight, does it not? A. It may be up to 50 per cent by weight.

Q. You speak here of production in a two-inch tube yielding about 65 per cent ethylene oxide. You mean that was the efficiency? What we have termed efficiency in the testimony? A. That is what we call yield and I believe that is what you term efficiency.

Q. And you say throughout the year 1936 and up to the middle of May, 1937, you carried out extended investigation, and so forth. In all that work was water present in the reaction zone? A. Water was not added.

Q. Answer the question, please. A. Water was present to the extent that it was in the air, which was fed to the apparatus.

Q. And to the extent also that it was formed in the reaction zone? A. That is correct.

Q. Is Dr. Metzger the technical director of the defendant? A. Yes.

Q. And he was acquainted with all your work during this time and was in a position where he perhaps best knew the program? A. Well, he was acquainted with the work, yes.

Q. And he has been in court throughout the trial? A. Yes * * * [1317-1328].

* * * * * * *

TESTIMONY OF CHARLES E. ADAMS.

Thereupon——

CHARLES E. ADAMS, a witness on behalf of the defendant, having been first duly sworn according to law, was examined and testified as follows:

Direct Examination.

By Mr. Edmonds:

Q. What is your place of residence and your official capacity? A. 4 East 72nd Street, New York City. In connection with this suit I am Chairman of the Board of the U. S. Industrial Alcohol Company and the U. S. Industrial Chemicals Corporation, Inc.

Q. What is the relation of those two companies? A. The U. S. Industrial Chemicals, Incorporated, is a wholly owned subsidiary of the U. S. Industrial Alcohol Company.

Q. What is briefly the reason for having the two separated? Is it something that grows out of the— A. Something that grows out of the structure of the two companies. The Alcohol Company started as a distiller of industrial alcohol. There are any number of restrictions surrounding an alcohol plant. The Government has inspectors all over the place. You are confined in your activities, so we formed the chemical company and brought the alcohol around to the chemical company to make further derivatives and then there were certain tax reasons for changing the capital structure and finally

there developed from that the U. S. Industrial Alcohol and the U. S. Industrial Chemicals, which is a selling and a chemical manufacturing company——

Q. Now, will you state what is the nature of the business of the alcohol company and what it was in 1904 and in that connection you may refer, if you will, to the use of ethylene glycol as an anti-freeze? A. The U. S. Industrial Alcohol Company started as a manufacturer of industrial alcohol. At the time industrial alcohol was declared tax free as distinguished from alcohol. Out of that developed the chemical enterprise which was based on the derivatives of alcohol in the beginning but over the years from 1904 to 1927 when I became connected with the U. S. Industrial Alcohol Company and from then on to the present, a very large proportion of their alcohol business was in the anti-freeze field. That is, ethyl alcohol was used as an anti-freeze medium in automobile radiators in the cold weather. From the time when I became intimately acquainted with the business in 1927, I would say 40% of the volume of industrial alcohol made by the company went into automobile radiators as an anti-freeze.

In 1927 the production and sale of ethylene glycol as a so-called permanent anti-freeze was relatively unimportant. It was just beginning. By the time 1933 rolled around our best observation was that ethylene glycol had taken about 14% of the business away from ethyl alcohol. We were obviously very much interested in having an ethylene glycol supply of our own to supplement ethyl alcohol as an anti-freeze. Ethyl alcohol will always have a big volume because it is a cheaper product and the man that hasn't the money to pay to charge his radiator with ethylene glycol in the beginning will take a chance that he will not have to charge it too many times with ethyl alcohol and that is always going to be a big volume, but we want them both. Do you want me to go on from there with the history of our development?

Q. Yes, if you will. A. Toward the end of 1933 we had a survey brought right up to date as to the proportion of business then enjoyed by ethylene glycol. We had a report made by one of our research men and that report indicated that there was no reason why we could not

develop a perfectly workable process for making ethylene glycol by the old fashioned chlorine method, but advised us very strongly against that on the ground that for twelve or fifteen years people had been working toward the direct oxidation of ethylene, and if we had a direct chlorine supply of our own and transferred the chlorine from its source of manufacture down to our plant, and made ethylene glycol, we would have an investment in what he told us would be an obsolete plant in the course of the next five or six years. So we determined at that time to start a real research program on the direct oxidation of ethylene and started spending money with that in view in January, 1934.

Q. What was the program initiated at that time and how did you follow through? A. That research problem was, as usual, given a number and assigned to the laboratory at Stamford, where we have some fifty-five research chemists in the employ of the Air Reduction Company and U. S. Industrial Alcohol. The expenses of that laboratory are borne jointly on a pro rata basis as indicated by the work done by each company over a period of time. It might have changed from time to time but it runs pretty even for forty per cent of the expenses going to the Alcohol Company and sixty per cent going to the Air Reduction Company.

Orders were given to throw the throttle wide open with full steam ahead to solve this direct oxidation of ethylene as soon as possible. Some of the best men we had were assigned specifically to that work and from that time on until we started construction of our commercial plant in midsummer of 1938 approximately, they were never off from that subject.

Q. At an early stage on that, did you do anything in the way of arranging to keep a supply of current literature of the subject coming in? A. It was already automatic, beginning in 1916 when the Air Reduction Company was formed and we started the Research Department, such machinery was set up so that every current chemical paper was brought to the attention of our experts and they followed all the patent applications from all the countries that have patent laws and received the reports and patents as issued, so that anything that we

were interested in, anything that was a related research problem or that the librarian, we will call her. in charge of that particular bureau, thought was related to what we were interested in, that was immediately sent for, brought in and read and turned over to the research man who was most closely related to that subject.

Q. Did the operation of that established system result in bringing before your people the French Lefort patents? A. Yes, indeed.

Q. Were they examined at the time? A. Yes.

Q. Did anybody find anything of interest in them? A. Not at all. That was reported in February, 1934, as I remember the first one.

Q. And the others came along? A. The others came along just as soon as they were issued.

Q. Will you trace the course of the research work, what was done, how it progressed, with what vigor it was advanced, and the period of time it covered? A. Well, it was pressed with all the vigor that they could press it. It had its up and downs like all research problems do, all which I have ever had any experience with. It was not until the end of 1935 that we began to get pretty well encouraged. Somewhere in the very early spring of 1936 we had achieved such results that we were in our own minds, I say in our minds, I had no mind on the subject, it was the research men that had the minds, but in their minds they felt we could sit down with patent counsel and outline the road that we expected to pursue and the goal that we hoped to reach as the result of following that.

Q. Did the research work cover research in both catalytic and non catalytic processes? A. Oh, yes, it covered them.

Q. Did the research cover catalytic operations, both with and without water? A. Yes.

Q. Can you state approximately, or at least by way of limitation at what time you had definitely discarded the idea of admitting the water to the reaction chamber, and from which time you went on using only the ethylene

and air reactions? A. I can spot that, yes. It was towards the end of—it was pretty close to the end of 1935 that we knew that we did not want water. Having taken that step, it led us to the indication to the road that I just spoke of, in the spring of 1936.

Q. Do you know whether the experimentation included experimentation with various different metals as catalysts? A. I know it included lots of catalysts, but you are getting a little out of my depth.

Q. Do you know whether or not it included— A. There are men here that can answer that question.

Q. Did it include a wide scope on work with different carriers for catalysts? A. Yes, I heard a good deal about various carriers, but I can not describe them.

Q. Did you come to know anything about the habit of a catalyst, as to how long it would last? Was its life an item of any consequence? A. Well, they were doing business, I know, with a silver catalyst. I was very interested every time we had a meeting, and I had a report as to how long the thing was going to last, and whether it could be rejuvenated, because it is a very considerable item of expense before you get through with it.

Q. I did not make my question very clear. Was the period of time over which a catalyst, whatever it was, would continue to act catalytically an item of importance? A. Yes; that is fundamental, I think.

Q. Some of them subside more or less quickly? A. I am sure there were such cases reported.

Q. Yes. Now, can you state, or will you please state as well as you can, when it was that you had arrived at what you considered to be a commercially operative process? A. In somewhere, I would say just after the middle, or just before the middle, I guess, of 1937, we decided that our experiments, except for enlargement, were practically over, and it was time to build a large pilot plant.

Q. My question was directed more particularly to when you had concluded in your own mind that you had arrived at a process that was commercial, not the engineering of

it, or the building. A. That was some time very late in '36.

Q. Now, by the spring of '37, had the engineering form of the apparatus taken definite form? A. As far as the pilot plant was concerned, yes.

Q. At that time did you feel that you had not only developed a process, a commercial process, but that you had developed one that involved invention? A. Yes.

Q. And did you apply for a patent on that invention? A. Mr. Carter applied for a patent.

Q. Is this the patent to which you refer, No. 2125333 (handing patent to witness)? A. The method and catalyst patent?

Q. Yes * * * [1277-1288].

* * * Mr. Watson: I will stipulate that such a patent issued to the defendant, filed so and so, dated so and so, number so and so. Beyond that I won't * * * [1292].

* * * The Court: Now, all you need to do is to give the reference to it, and the issuance date. I will limit it to that. In other words, it is a reference in the course of his chronological statement, and that is all.

Mr. Edmonds: Yes; and the reference is by stipulation to Patent No. 2125333, issued August, 1938, on an application filed May 8, 1937; and it was issued to the U. S. Industrial Alcohol Company as assignee of R. M. Carter.

The Court: Let me see if I understand it. Doesn't Mr. Adams' testimony boil down to this—I do not want to anticipate too much—I understand this would be conceded, that his company has built up a very large business in connection with this process.

Q. (Mr. Edmonds) Will you proceed to tell the rest of the story from where you are now? We will say you are at this application date, May 8, 1937, did the engineering development work continue from that time on? A. That May 8, 1937, marked the approximate date of

the completion of our experiments in, we will say, a single tube test, a single tube operation * * * [1292-1293].

* * * Q. Did you continue with the engineering developments from that time on? A. From that time on we went into a very considerable pilot plant design, construction and operation.

Q. Can you tell us approximately when that was built? A. The engineering of it was started about that time. The plant was—the engineering started on the pilot plant a little before that. The pilot plant, if I remember correctly, was in operation just before or just after the middle of the year 1937: It continued to operate, with certain ups and downs, certain measles and chickenpox and children's diseases, but with increasingly satisfactory results, so that we immediately started the engineering for a commercial unit. And by the time that we had, we will say, run the pilot plant for nine months the engineering was complete on the commercial unit, and we, in the middle of 1938, began the construction of the commercial unit.

Q. And that was completed, and the plant put in operation— A. And that was completed and put in operation in May, May 9, 1939. And it has been operating very satisfactorily ever since.

Q. Here in Baltimore? A. Here in Curtis Bay, Fairfield, just outside of Baltimore.

Q. Can you tell us what was the total cost of the development work that you went through in this research and development that you have told us about, up to the date September 25, 1936, that being the date of the application for the reissue? A. I have had those figures broken down, and that was $132,000, approximately.

Q. That is up to September 25, 1936? A. '36.

Q. Now, it has been continued from that date up to May 18, 1937. And I wish you would state that figure. A. I have those accounts to show that $19,000 more was spent before the May date.

Mr. Edmonds: That is all.

CROSS EXAMINATION.

By Mr. Watson:

Q. Early in May, 1939, this interrogatory was propounded by plaintiff to the defendant: "Since May 18, 1937, and prior to the filing of the complaint herein, has defendant, or any corporation in which defendant at the time owned and now owns a controlling stock interest, commercially practiced a process of making ethylene oxide by so subjecting a mixture containing ethylene and molecular oxygen at temperatures maintained between about 150 degrees C. and about 400 degrees C. to contact with an active surface catalyst comprising silver for direct oxidation of ethylene by molecular oxygen as to form oxidation products containing substantial amounts of ethylene oxide and not containing substantial amounts of aldehydes, and thereafter recovering ethylene oxide. (By 'commercially practiced' is meant—conducted on a commercial scale in the regular course of business and producing a product or products for the purpose of sale and sold in the original channels of trade, as distinguished from experimental or laboratory operations.)"

Now, you were advised of that interrogatory about that time? A. Yes.

Q. And it was answered, as you know, No, is that correct? A. I don't know about the date.

Mr. Edmonds: Mr. Watson, did you say that was in this suit?

Mr. Watson: Your Honor may or may not recall that a bill of complaint on the patent in suit was filed against the U. S. Industrial Alcohol Company. This interrogatory was asked in that suit. It was answered negatively. And on the basis of that negative answer the suit was discontinued. A month or two later the present bill of complaint was filed against the U. S. Industrial Chemicals Company. And that is the present suit. But this interrogatory covers the operations of the present defendant, as well as the operations of the Alcohol Company. And I want it on record that we have a sworn answer, a sworn statement on behalf of the Alcohol Company, which wholly owns the subsidiary, the defendant here, that prior

to May 3, 1939, neither of them had ever made and sold a drop of ethylene oxide or glycol produced by this process.

The Court: Well, now, in that connection I would like to ask the witness a question. I understood you to say that you did not get into commercial production until considerably after the date of this reissue patent, namely, considerably after May 18, 1937.

The Witness: My recollection is it was in May, 1939, that we got into production.

The Court: Two years later?

The Witness: Yes, sir.

The Court: Now, after this patent was issued your company presumed that you knew all about it?

The Witness: We did.

The Court: Did you have correspondence with the plaintiff company, trying to find out what each was doing?

The Witness: No.

The Court: There was no correspondence——

The Witness: No.

The Court: —in regard to possible conflicting rights.

The Witness: No. We felt sure that we did not conflict.

The Court: You went ahead?

The Witness: We obtained advice and decided it did not conflict.

May I say a word of explanation, to simplify this last matter that came up?

The Court: Yes.

The Witness: When that interrogatory came in and reached my desk, I called up the President of the Carbide Chemicals Corporation, and told him that they had asked the wrong company, and that they should ask the operating company, the U. S. Industrial Chemicals, Inc.

Q. (By Mr. Watson) Now, Mr. Adams, perhaps your memory is at fault. Didn't you tell Mr. Rafferty that the Chemicals Company at that time had not commenced commercial operations? A. Maybe I did. But I said they would commence in a few days, we expected. That is possible. But it would be the Chemicals Company that did it.

Q. Yes. A. I told him that.

Q. So that the Chemicals Company as of that date had not begun any commercial operations; isn't that right? A. There may have been a few days there. I told him we would let him know exactly as soon as the thing began operation. That is exactly what I told him.

Q. And then you did not tell them— A. I told him as soon as it began commercial operation I would let him know. You can not tell me I did not tell him, because I did, and he will admit it * * * [1294-1300].

* * * Q. Mr. Adams, you spoke of being advised by your counsel that you could freely make glycol by the chlorhydrin process. That was about 1934? A. That was at the end of 1933, but, as my testimony will show——

Q. But you were advised, as far as patent questions are concerned, that was free? A. That a process could be devised under that chlorhydrin method that would undoubtedly be free from patent infringement.

Q. You were advised in about April or May of 1936 that plaintiff here had acquired the United States patent rights to the Lefort process? A. Yes.

Q. And you also knew before you received that information that your company had been in correspondence with the French company regarding the American patent rights? A. No, not regarding the American patent rights, regarding the amount of knowledge that they had of this art.

Mr. Watson: I hand you a folder of documents which concern communications back and forth between you and Mr. Munson and Mr. Board and the French company and the Guaranty Trust Company in Paris. These

we have been given by defendant's counsel. I offer them in evidence as a group as Plaintiff's Exhibit 22. They speak for themselves.

(Photostat copies of correspondence between defendant company and its officials and French company and Guaranty Trust Company in Paris, received in evidence and marked, "Plaintiff's Exhibit 22.")

Q. You spoke of the expense involved in the use of a silver catalyst. You realize, do you not, that the silver used and the expense of the silver was not ephemeral, the silver did not run away from you? A. Yes, of course.

Q. You could always get your money back out of the silver? A. We used silver because it happened to be in this catalyst.

Q. Yes, but it was not consumed in the process? A. Oh, no.

Q. You could cash it in at any time it failed? A. But the life of the catalyst was what he was asking me about, and the interruption of the production is very expensive.

Q. You spoke of the semi-pilot or pilot plant which you said went into operation about August, 1937— A. Somewhere about the middle of 1937.

Q. 1938, I guess it was? A. No, 1938 is when we started the commercial plant.

Q. 1938 is when you started to build the commercial plant? A. That's right.

Q. Which commercial plant you started to operate in 1939? A. That's right.

Q. You have spoken of the cost of your experimental program before November, 1936. You began communications with the French company in November or December, 1935, did you not? A. No, in February, 1936.

Q. Well, you will accept whatever date is shown by the documents just offered in evidence? A. Well, it is February, 1936.

Q. And you are familiar with the fact, are you not, that up until that time, so far as the evidence in this case shows, your company or any of your subsidiaries had done no work, even on a laboratory scale, on the production of ethylene oxide or by direct combination of ethylene and molecular oxygen from air, except Mr. Carter's February, 1935, couple of tests when he ran a gas mixture over a five cent piece size of silver gauze, and in December of that year when twice he attempted to pass it over silver rivets? A. I can not answer that.

Q. Are you familiar with that? A. I am not at all.

Mr. Watson: That's all.

REDIRECT EXAMINATION.

By Mr. Edmonds:

Q. Prior to writing this letter of February 26, 1936, had you had any interest whatever in the original Lefort patent, which by that time had been issued? A. No, none whatever.

Q. Did you ever ask anybody in the Patent business to tell you anything about it? A. No, not up to that date.

Q. Prior to this letter of April 29, 1936, had you gotten a report on the patents in this field of ethylene oxide? A. I had.

Q. Did that include any note about this Lefort patent? A. It certainly did.

Q. Did it include anything definite to you about whether or not the patent was of any value to you? A. They said, under the circumstances——

Mr. Watson: That is hardly the best evidence.

The Witness: Under the circumstances that what you are aiming at, that patent is of no interest to you.

Mr. Edmonds: That's all.

The Witness: That was April 14, 1936 • • • [1304-1309].

TESTIMONY OF WALTER WILKINSON

Thereupon——

WALTER WILKINSON, a witness of lawful age, produced on behalf of the defendant, in surrebuttal, having been first duly sworn, was examined and testified as follows:

Direct Examination.

By Mr. Edmonds:

Q. You are in the employ of what company? A. The Air Reduction Company, Incorporated.

Q. And in what capacity? A. Research engineer.

Q. And have you been identified with the ethylene oxide development of the U. S. Industrial Chemical Company during the past several years? A. I have.

The Court: I do not understand what you mean.

The Witness: I am the research engineer of the Air Reduction Company, Incorporated.

Q. (By Mr. Edmonds) And what is the relation between that company and the U. S. Industrial Chemical Company? A. The Air Reduction Company, Incorporated, run a joint laboratory with the U. S. Industrial Chemical Company • • • [1015-1016].

• • • Q. (By Mr. Edmonds) You are a practical engineer, with some engineering knowledge? A. I am a graduate of Stevens Institute of Technology in 1917.

Q. You heard the testimony of Dr. Lamb? A. I have.

Q. I call your attention to the Court's question put to him on page 794, and his answer thereto, and also his answer to a closely related question on page 795, and ask you if you will state your views with respect to the matter treated in those two answers (handing transcript of testimony to witness). A. In the first question I take it Dr. Lamb was asked if the experiments which he had performed, in view of the yields which he had obtained, the conversion, would lead him to say that this was a commercial process, in view of these yields.

Q. Yes. A. In view of these yields or conversion, Dr. Lamb testified that it appeared to him to be a commercial process.

Q. That is solely in view of the conversion? A. Solely in view of that, as I take it from that question.

Q. Now, the second question has no such limitation, has it? A. In the second question I took it to be that the question referred to all points involved in the situation * * * [1016-1018].

* * * Mr. Edmonds: The second one reads, "In other words, if you were asked for such advice as I have just suggested, you would be willing, on the experiments that you have made and none other, and the study that you have made and none other, to give them an affirmative answer, Yes?" That is "Yes" as to its adaptability for profitable commercial successful operation.

Q. (By Mr. Edmonds) Now, do you agree with that statement? A. I do not.

Q. Will you state your reasons? A. There are many other things to consider in a commercial operation besides the conversion of ethylene to ethylene oxide.

The Court: Well, that is all we have been talking about.

The Witness: But there are many other things to consider besides the conversion, to make that a commercial operation. The length of life of the catalyst is one. And for short tests, over a period of a few minutes, you can not determine what the length of life of that catalyst may be. And if the length of life of the catalyst is not sufficiently long, the cost of operating that process would be too high, because you would have to do something about replacing or regenerating the catalyst. And, therefore, for that reason you can not from one experiment on one catalyst alone, with only a few analyses taken, over a period of a short time, you can not state that that would be a commercial procedure. You have not enough information to give you commercial procedure. You could not recommend, for instance, that you would spend hundreds of thousands of dollars to erect a big plant on the basis of that information

which you obtain from one small experiment, and consequently you would have to go back to the laboratory and ask the laboratory people to do more research work before an engineer could even get started on the problem. An engineer must know a good many things about a process before he can extrapolate it into a commercial proposition.

The Court: Well, what do your remarks mean? What do you mean to have inferred from your remarks, that the patent is no good, or that the patent is all right, but simply that Professor Lamb was talking too broadly? I do not see what——

The Witness: Professor Lamb was talking too broadly when he said that he could make a direct statement that from those experiments that he had performed it was a commercial process.

The Court: Let us assume that he did, where does it get us in the present case? We are concerned with whether or not the patent is valid, and more specifically with whether it gives a sufficient disclosure. And, as I understand it—maybe I do not fully understand it—I understand that what you mean to say is that you would not be quite so bold as Professor Lamb, you would want to do more. But if the patent tells you how to do more, and if you can find what we call a permanently good catalyst, then you might do the same thing that he said he would do more hastily. Isn't that about all it is?

The Witness: You might try to find a catalyst which you considered to be satisfactory for that purpose, for that procedure.

Q. (By Mr. Edmonds) Are you familiar with the developments of the U. S. Industrial Chemical Company through 1936, when this was going on? A. Yes, I am.

Q. Did the U. S. Industrial Chemical Company produce a catalyst having a pretty good rate of conversion? A. We did • • • [1018-1021].

• • • Q. What particular catalyst and support do you refer to in your last answer? A. The catalyst where the silver was deposited on pumice.

Q. Now, to go back to the first two questions asked Mr. Lamb, I ask you, if you assume that the rate of conversion that he was talking about there was the rate that would continue indefinitely unchanged, still would that be a fair basis for a recommendation that profitable commercial operations could be conducted in that way? A. In the first question of Dr. Lamb—— * * * [1022].

* * * The Court: What page are you reading from?

The Witness: I am not reading from any page.

Mr. Edmonds: The first question is 794. Your Honor's question you will see right at the end was limited to a statement based on the rate of conversion only. And I am asking him to limit himself, too, in that way to a catalyst which maintains that rate of conversion unchanged.

Q. (By Mr. Edmonds) Even so, would that experiment be a fair basis for a recommendation of the adoption of that method for profitable commercial operation? A. That is, if the pumice——

Q. Assuming that the rate of conversion lasts, without any deterioration? A. Well, we would have to consider a few other things in that.

Q. That is what I want you to mention. A. We would have to consider the fact that it is over a catalyst composed entirely of silver, which is an expensive catalyst.

Q. State what is involved in that, and how that affects this matter of profitable commercial operation. A. The cost of the catalyst is a very important thing in the total cost of the plant, provided it is a metal, like silver, which has a high value per pound. And if you were using a catalyst composed entirely of silver, and if the rates of production that you got, not merely conversion of ethylene, but the total ethylene oxide you got per pound of silver, if those things are low, then you must use large quantities of silver; and you might even use a large enough quantity to make the process uneconomical. That is why the use of supports is indicated, because by putting the silver on a support you will decrease the amount of silver and thereby decrease the total cost of the catalyst, if you can maintain the same yields and the same

total production per volume of catalyst with the catalyst on the support as you do with the catalyst using all silver.

Q. Using the catalyst referred to in that question and answer that we are talking about, would it be possible to conduct profitable commercial operations at the present day? A. I have seen nothing from the testimony of Dr. Lamb, or in his experiments which he has put in the record, which would indicate to me that you could take that particular catalyst and that particular type of equipment in which he tested it, and translate it into commercial operations, whereby you could make ethylene oxide at a profit.

Q. I think you limited that a little too closely. Take that general type of catalyst, relatively pure silver, or pure silver, whatever it was, what I am asking you is would that involve such a cost, such an investment, as to make it impossible to conduct profitable commercial operations on that basis? A. Well, if you just translate that amount of silver that Dr. Lamb used to a plant making, say, five million pounds of ethylene oxide per year, you would have, assuming that the silver had a density of about 1.3 compared to water, you would have approximately $300,000 worth of silver involved in the cost of the plant.

Q. And would that make the operation unprofitable, that amount of investment in the catalyst? A. I would say that would make it entirely unprofitable.

Mr. Edmonds: That is all [1022-1025].

CROSS EXAMINATION.

By Mr. Watson:

• • • Q. Have you ever conducted a commercial catalytic process? I am speaking of you personally? A. I have built, designed and built processes for the oxidation of such things as isopropyl alcohol into acetone, and ethyl alcohol into acetaldehyde: And these oxidations took place over a silver catalyst, with the addition of air and isopropyl alcohol or ethyl alcohol, as the case may be • • • [1026].

• • • • • • •

TESTIMONY OF ARTHUR B. LAMB

Thereupon——

DOCTOR ARTHUR B. LAMB, a witness of lawful age, produced on behalf of the plaintiff in rebuttal, having been first duly sworn according to law, was examined and testified as follows:

Direct Examination.

By Mr. Johnson:

Q. Will you tell us your age and occupation, Doctor Lamb? A. I am sixty years old. I reside in Brookline, Massachusetts, and I am professor of chemistry at Harvard University.

Q. Do you recall that in the latter part of April of this year I asked you to come down to my office in New York and, if so, will you tell us what occurred? A. I was called to your office in New York in the latter part of April, I think it was April 27th, and at that time you showed me in the presence of Mr. Watson the patent referred to as the original United States Patent of Lefort, which has been discussed in this case, and you asked me if with that document I would undertake to carry through the preparation of ethylene oxide and after examining the patent I agreed to undertake this test.

Q. Did you later do that? A. I did.

Q. Prior to making this test or speaking to me on the subject, had you read or were you informed of the contents of the Reissue patent of Lefort, which is the patent in suit? A. No, I did not see the Reissue patent prior to this conference, during it nor any time thereafter and I have never as a matter of fact read the Reissue patent.

Q. Will you tell us your education and experience which you consider qualify you to make such a test and testify thereon? A. After a considerable period spent in training and education, I took charge during the war of the Chemical Research on the defense side of chemical warfare and one of the problems that confronted us at that

time was the removal of carbon monoxide from air. It was in this occupation that I made the acquaintance of Doctor Frazer and became familiar with his great outstanding accomplishments in this connection. Other research groups were busied with this same problem and I was in charge of their operations and studies. After the Hopcalite catalyst had been perfected and its efficacy demonstrated, it was necessary to manufacture it on a considerable scale to be enclosed in cannisters that could be used in connection with masks by the soldiers and sailors in our forces to guard them against carbon monoxide poisoning. A plant was constructed and many tons or several tons of this material was manufactured under my general direction and some 30,000 such cannisters were filled for the United States Navy and were tested and shown to be efficacious as a protection against carbon monoxide.

After the war I was asked to organize and to direct the Fixed Nitrogen Research Laboratory in Washington, established by President Wilson for the purpose of, if possible, converting the immense Government plant at Muscle Shoals into effective plants for the manufacture of fixed nitrogen from the air. Our chief problem and the one to which we devoted the most time and money was the development of a catalyst for the production of ammonia from hydrogen and nitrogen. After a considerable inspection trip in Europe and after some months, more than a year of effort, such a catalyst was actually developed and has played a considerable role in the subsequent manufacture of ammonia in this country.

Subsequent to my connection with the Chemical Warfare Service and the Fixed Nitrogen Laboratory, I carried on some researches particularly with regard to the behavior of Hopcalite towards carbon monoxide and hydrogen under different conditions. This, I think, completes my active connection with catalyst research.

Q. What about your education, you haven't mentioned that? A. I studied originally at Tufts College in Massachusetts and then at Harvard University and received from both places the degrees of Doctor of Philosophy. I have studied in several universities in Germany and returned to this country to become an instructor in chemistry at Harvard University.

Q. If it won't embarrass you, will you tell us what honors you have received in the scientific world? A. Well, I have received more than I deserved. I have been elected to some honorary societies and received honorary degrees. I have been president of the American Chemical Society.

Q. In fact, you are the editor of the journal of that society, aren't you? A. I am and have been for over twenty years.

Q. And you are a member of the National Academy of Sciences? A. Yes.

Q. Did you prior to making this test on the original Lefort patent receive any suggestions or instructions from me or anyone as to how you should conduct this test? A. No, sir, with this exception, I think you or Mr. Watson did tell me to remember that I was not expected to produce this material on a large scale, that a laboratory experiment would be adequate.

Q. Will you please tell us, therefore, what you did and what results you obtained? A. May I refer to aid my memory to a report which I have recently written outlining this work briefly?

Q. Yes, perhaps you had better begin with the apparatus. Did you make a drawing of it or did you have one made? A. Yes, I made a drawing of the essential part of the apparatus, the catalyst cell, that is, a glass vessel containing the catalyst.

Q. I hand you a drawing and ask if you will identify it. Is that it? A. Yes, that is a diagram of this apparatus drawn approximately to scale.

Mr. Johnson: I offer the diagram identified by the witness as Plaintiff's Exhibit 11.

(Diagram of Doctor Lamb marked "Plaintiff's Exhibit 11.")

The Witness: The apparatus to contain the catalyst was a simple one, merely a small glass tube held in a vertical position in which the catalyst was placed and into which a thermometer was inserted, whose bulb reached into the catalyst mass. The cell was partially

submerged in a liquid bath of dibutyl phthalate, so that it could be heated to a moderate temperature when desired. In order to be sure that the gas mixture entering the cell had reached the temperature of the catalyst, we have some spiral coil glass tube through which the entering gas has to flow before it strikes the catalyst. Passing through these glass tubes warms the gas mixture sufficiently to the temperature of the liquid bath in which the cell is placed. The liquid bath is heated on the bottom by a little hot electric plate or electric heater to a convenient temperature. The apparatus for preparing the mixture of ethylene and air followed conventional lines. The ethylene was supplied from a pressure cylinder and then passed through a pressure regulator so that any excess of ethylene above the desired pressure would escape. The ethylene then passed from there through a valve and a flow meter to measure the rate of flow and then into the mixing chamber just in advance of the catalyst cell. Air was taken from a compressed air line of the laboratory, bubbled through the bottle filled with water with a trace of sulphuric acid in it. The function of the bottle I will explain later. Then, I should like to correct that—the compressed air passed first through a pressure regulator so that any excess pressure could be removed and then the air at reasonably constant pressure entered this wash bottle containing the water with a drop of two of sulphuric acid solution to which I have referred. From the wash bottle the air passes through a valve, a flow meter and into the mixing chamber referred to before. By this preliminary apparatus we were able to supply to the catalyst itself a current of air with a regulated amount of ethylene and containing small amounts of water substantially equivalent to that present in ordinary atmospheric air. In order to detect the ethylene oxide, if any, in the gases issuing from the catalyst cell, we bubbled those gases through solutions first of salt and water, to which a little hydrochloric acid had been added. In some of our experiments we also added so-called wash bottles containing some sodium hydroxide solution to collect at least a part of the CO_2, that is carbon dioxide, that might be formed in this catalyst cell. In other experiments we also collected the carbon dioxide completely by absorbing it in so-called U tubes charged with pellets of fused potassium hy-

droxide. Those were our general experimental arrangements.

Q. And did you in making this apparatus or in the tests which you subsequently conducted on it, use consciously any knowledge that was not available to the art in 1931? A. No, I didn't.

Q. Now, will you tell about the preparation of your catalyst and what you did. Perhaps this will be shortened, your Honor, if he has—he has a report which we could put in.

The Court: I think you might just let the professor go ahead and describe it.

The Witness: I first read the patent with a good deal of attention and devoted some little thought to the question of the selection of a catalyst because I felt some little professional pride in being able to pick out, if possible, a catalyst which would work the first time. At least, I did not want to make too many trys. I reasoned that the most promising single catalyst would be that comprised or consisting solely of silver because silver was so frequently mentioned in the patent and I reasoned that would be about the simplest one to prepare. I further reasoned that it was clear that a quite active catalyst was desirable because we wanted to accelerate a reaction at a low temperature and avoid a reaction which I knew took place with greater facility at high temperature.

The oxidation of the ethylene to carbon dioxide and water is one that would take place freely at high temperatures. The oxidation of the ethylene to ethylene oxide, according to the patent, took place at low temperatures, particularly around 150 to 400 degrees, and I could see that it was desirable to have the silver in as active a form as I could get it and I knew that this meant fine subdivision. I therefore decided that I would utilize some compound of silver that would decompose at a very low temperature, certainly if possible at a temperature no higher than the range specified in the patent for the use of the catalyst and I knew that silver oxide fitted those requirements, so I prepared silver oxide from silver nitrate by the regular methods of precipitating with sodium hydroxide and washing out the impurities left in the preparation as carefully as I could by repeated

washing of the silver oxide by what we speak of as decantation, pouring off the wash waters each time, in what is known as centrifuging the precipitate with a little of the wash water remaining so as to get as clean a silver oxide as possible.

We repeated this process a good many times until we were assured that the silver oxide was reasonably pure. I then had that baked in an oven to dry it at 115 degrees over night and in this way I got a cake of silver oxide about four inches in diameter, perhaps 4 inch diameter, but it was quite firm and hard. I then had that broken up carefully into granules so as not to get any more fines than possible because I did not want to waste the silver and wanted to have a granular product. This was carefully done. As a matter of fact, we only got a few fines and obtained a granular silver oxide about the size of a BB shot and free from fines.

We placed those granules of silver oxide in a glass tube, about eighty grams of them, and then proceeded to decompose that silver oxide in a furnace at a carefully controlled temperature. We plunged a thermometer right into the silver oxide and also into the surrounding furnace and raised the temperatures very slowly and then attached a device to the end of it, a glass tube, so we could tell when the decomposition of the silver oxide was progressing rapidly, and when it was complete, that is, when no more oxygen was given off, and in this way we were able to decompose all of this silver oxide with reasonable rapidity. I think it only took us a couple of hours at temperatures that never exceeded 390 degrees Centigrade.

You see, I was anxious to keep below the limiting temperature of 400 degrees given in this patent because that would be the working range of this catalyst when we got it, in this heating of silver oxide granules, these granules of silver. We were delighted to find that they were quite firm granules free from fines and we examined them under a microscope and they were very porous, at least very fine grained and were very light colored; they looked good.

We put them in our apparatus and proceeded to make the experiment in our catalyst cell. Of course, I didn't

know whether we would get any ethylene oxide or not, so I did not attempt in the first experiment to collect any of it. I simply wanted to prove its presence, so that I took some of the absorbing solution with just a few drops of acid in it and some indicator, methyl red, which would turn from red to yellow when all the acid was used up. The ethylene oxide uses up the acid and so if the acid was used up, I knew I had ethylene oxide and so you see now the reason for the wash bottle I put in at the start. I put the wash bottle in with water containing a few drops of acid so as not to have any ammonia or other alkaline ingredient which might vitiate my test and fool me and make me think I had ethylene oxide when I did not. This slightly acidulated water prevented me from making this error and I was naturally delighted after three minutes when I got a switch from the red to the yellow at a temperature of 190 degrees in the catalyst bed and at that same temperature I could see a slight differential in the reading of the two thermometers, the thermometer in the catalyst was a little hotter, two or three degrees hotter, than the thermometer in the bath outside. That was a good sign too, and then I let the temperature rise higher and higher. The differential in the temperature indicating a very rapid formation of ethylene oxide and, surely enough, additional amounts of acid added to my absorber were consumed in that first run. I added again tenth molar acid and had it all changed over, showing ethylene oxide had been formed to use up all that acid. That was the first experiment, and then we knew we had gotten ethylene oxide.

Subsequent experiments were designed to determine the amounts more accurately that were formed and I have a table here that gives a summary of the per cent. that were obtained under different conditions.

In some 11 experiments all told the period of collection in these experiments varied from 15 minutes to 30 minutes; and in the first experiment, for instance, I got 28.7 per cent. of the ethylene converted to ethylene oxide. Out of a mixture containing ten cubic centimeters of ethylene for every 510 cubic centimeters of air added.

The Court: What, in your opinion, is a good percentage of conversion?

A. Well, to tell you the truth I did not know at the time, but I have since learned that this is a good conversion.

This is a yield of ethylene oxide from ethylene. My wash bottles showed some CO_2 formed, and while I did not know exactly how much this was, I knew it was considerable, but to test this I made a final experiment where I collected all of the CO_2 formed, and in this particular experiment I got 37.7 per cent. yield or conversion of ethylene to ethylene oxide, and I got a 43.7 conversion of ethylene into carbon dioxide, or about the same volume efficiency in both of these directions.

I do not think I told you perhaps the one essential point you should know——

The Court: What about the carrier? Did you mention that?

A. I did not use any carrier. There, again, I did quite a little thinking. I realized that I had not much time, and I knew that some catalyst carriers were active in decomposing ethylene oxide, and I said, "Why assume this handicap? Why not just use straight silver? Then I don't have any worry over the catalyst carrier." • • • [755-776].

• • • Q. This table you have identified is page 4 of the report which you have in your hand? A. Yes.

Q. Will you state the efficiency of that last run where you actually measured the CO_2 carefully? A. I did not state it, but——

Q. Can you calculate it? A. I can calculate it. (Calculating on paper.)

The Witness: The efficiency defined as the per cent. of the ethylene converted to ethylene oxide based on the total ethylene consumed in passing through the catalyst, is 46 per cent. in this last experiment.

Q. And the conversion percentage is what? So that we will have them right together. A. 37.7 per cent. conversion of ethylene to ethylene oxide and 43.7 per cent. conversion of ethylene to carbon dioxide.

The volume of the catalyst used in this little cell was only 17 cubic centimeters. The temperature in this last experiment throughout the period of collection in the catalsyt bed was 230.3 degrees Centigrade.

Q. Will you define the efficiency again, what you mean by that? The amount of ethylene consumed divided into what? I believe you did not state it in your previous answer. A. I added together the percentages of the ethylene converted into ethylene oxide and into carbon dioxide, and I divided the percentage of ethylene oxide formed by the sum of these two percentages and multiplied by 100 per cent. and obtained the answer given.

Q. All right. Now, you have stated that you passed the air used through this wash bottle, not only to be sure that no ammonia or other alkaline material went into the reaction zone but also to insure that the air was approximately normal atmospheric air, as I understand you. Do you believe, from your study of the Lefort original patent that I gave you, that water is a necessary requirement in the process to produce ethylene oxide? That is, large amounts of added water? A. From my reading of the patent I had understood that water was, in general, beneficial to this production of ethylene oxide. It was given in most of the examples, and it was recommended to promote the production of ethylene oxide and I thought as long as I was anxious to get some, I had better have a little water in there, so I did not dry my air. I decided to use it either as it came or with the slight additional amount of moisture that it might pick up from going through the wash bottle, because I knew that the air that would go in from the compressor line was somewhat less humid than the original outside air before compression.

Q. But, as to further added amounts, do you think that the Lefort patent requires that? A. No, it never occurred to me that it was required to add any water to the mixture, but that it would be a pretty good thing to do it, in view of the aggregate that was said to facilitate the reaction.

Q. How long did you run this apparatus in your tests, approximately? A. Well, it took quite a long time on

each run because I wanted always, before I began my period of collection I wanted to have the conditions pretty accurately stabilized, so I would keep the current of air and ethylene gas passing through the catalyst cell and make readings of the temperature in the cell, sometimes for hours, before I got a good steady state so that I could know within a degree the average temperature of the catalyst mass during that 15 or 30 minute run. So that this catalyst, which I have told you about, was operated a good many hours and produced a lot of ethylene oxide, considering its size.

Q. And during those tests did you change the water in the wash bottle? A. No.

Q. Did you notice how much it went down, and from that will you tell us approximately how much water you put in? A. Yes, I made a rough estimate of how much water that wash bottle had lost in the 11 or 12 runs that were made, because I knew how full it was at the start, and measured down on it, and I could guess about how much had been lost. It was a Friedrichs wash bottle of the blown type with an annular spiral space just inside of the wall of the wash bottle so that the cross sectional area was not very great, and I estimated it was less than 10 centimeters of water that had disappeared in that period.

Q. In how long? A. Well, about 50 hours of total operation.

Q. And at what rate were you passing air and ethylene mixture through there? A. Well, I can't tell you exactly because I know I passed it a half a liter a minute through the catalyst, but I only used part of the gas or air from the pressure regulator for that purpose. It spilled a good deal of the air without ever sending it through the catalyst cell, so that the total volume of air that went through the wash bottle must have been on the order of a liter and a half a minute, or, perhaps, say, 900 liters an hour. That would be some 30,000 liters all told.

Q. And what percentage of moisture in air would that calculate to approximately?

* * * Moisture in the gas mixture, I should say. I believe you said a while ago 1½ liters per minute was 900 liters per hour. That is not correct, is it? A. Well, I think I better calculate it. (Calculation made by witness.) An approximate calculation shows that this would have added about .2 of one per cent. of moisture to the air that flowed through the wash bottle.

Q. Now, from this first test which you ran in this small apparatus, and having obtained ethylene oxide in the amounts you have said, what did you conclude as to the sufficiency of the disclosure of the Lefort patent to produce ethylene oxide? A. Well, I concluded, of course, that it was adequate.

Q. You also called my attention to the fact that you ran another test, on which you used a carrier; is that correct? A. Yes.

Q. Will you speak of that? A. After I had succeeded so well on this first one, I thought I would now try one with a carrier. And casting about it seemed to me that ordinary earthenware offered the best hope for an inert carrier. Of course, the advantage of a carrier was to curtail the use of silver. So we prepared granules of unglazed earthenware, breaking up the ordinary porous plates that we use in the laboratory into granules about the size of BB shot, as before, and then we impregnated these granules with a silver nitrate solution, and quite a concentrated silver nitrate solution, and evaporated and drained, and then undertook to decompose the silver nitrate thermally at low temperatures; but we found we could not decompose that material at as low temperatures as had been efficacious with the silver oxide. Well, the best we could do was a prolonged heating at about 450 degrees Centigrade, pumping out the oxides of nitrogen occasionally, and continuing the heating for a good many hours, and ultimately got out, as far as we could tell, all the oxides of nitrogen, and having nothing but metallic silver left behind. We examined the earthenware granules under the microscope, and found that we did have metallic silver partially encrusted on the grains, and not completely covering the whole grain, but the silver did not have the mat appearance. It was a shinier silver, and did not look so porous. So we were a little pessimistic about how it would work. We put it in the cell,

the catalyst cell, and repeated our tests, and found a very slight activity in this sample.

Q. Up to what temperature? A. We got no noticeable temperature rise at 190 degrees. When the temperature reached 245 degrees we did notice a little differential temperature, four or five degrees differential, and then we did begin to get a little ethylene oxide. We kept running it thirty minutes, because of the small amount. We got .9 of a per cent. formation of ethylene oxide and some carbon dioxide. We did not know just how much, but approximately all we collected in this apparatus was a quarter of a per cent. carbon dioxide. So our actual tests confirmed our pessimism; and we decided that it was necessary to make another experiment, that we would have to decompose that silver nitrate at a lower temperature, probably by careful reduction in hydrogen. But we did not do that at that time.

Q. What was the temperature limit of the bath which you were using? Could you have carried that to much higher temperatures than that? A. Well, somewhat. We used dibutyl phthalate for the bath, and at 250 degrees it began to smoke a good deal, and decompose, so it would not be feasible in this bath to go to a much higher temperature, and we would have had to alter our experimental arrangements.

Q. Did the results which you obtained on this second test affect your opinion previously expressed as to the sufficiency of the disclosure of the original Lefort patent? A. No, it did not change my opinion.

Mr. Johnson: I offer the written report which the witness has identified as Plaintiff's Exhibit 12.

(Report of Professor Lamb marked "Plaintiff's Exhibit No. 12.")

Q. (By Mr. Johnson) You also made a curve, which I have on graph paper. What does that show (handing paper to witness)? A. Well, this gives in graphic form the percentages of ethylene oxide obtained in our several experiments. I had expected to make a better job of this, from the draftsman's point of view, but I just did not have time. But it shows that as the temperature rises, from the lowest temperature we worked at, 215 de-

grees, up to the highest temperature of 245 degrees, there is a gradual rise in the amount of ethylene oxide produced. The curve seemed, however, to be approaching a maximum. I don't know how much higher it would rise.

Mr. Johnson: I offer the graph which the witness has identified as Plaintiff's Exhibit 13.

(Graph prepared by Professor Lamb marked "Plaintiff's Exhibit No. 13.")

The Court: I have one or two questions, Prof. Lamb, that I would like to ask.

You said that you did not consider that water was mandatory, that is, the addition of water was mandatory in the patent. In that connection, I would like to ask you how you reconcile that conclusion with the fact that all of the claims, that is 1 to 7, inclusive, speak of a process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, or of oxygen and steam. Do you reconcile it by the fact that you think that may be taken as referring to the water that is present anyway in the atmosphere, in the air used?

The Witness: Well, I thought that, too. And then I thought, also, that the patentee had claimed what he thought was his best embodiment of the invention, which was having the water in it. There seems to be no doubt in the specification that water was beneficial. I think he claimed just that. I did not really study the legal aspects of it at all. But——

The Court: No, I did not intend to ask you from the legal aspect. Then, it would necessarily follow, I assume, from your conclusion, that even though, as I understood you to say, you have not up to the present time read the reissue patent, it would follow from what you have said that even if you studied it, and even if it be assumed that water as a requirement, we will say, is left out of that, you think that is not any material departure, not in any legal sense, but in a practical chemical sense.

The Witness: Yes, that is true, your Honor. As a matter of fact, when we first constructed the apparatus, an hour before we started our run we did not have any

wash bottle in there. It was not until I thought, that it occurred to me, just as we were going to start the experiment, that, "See here, how do I know there is not a little ammonia in my air that I am drawing in from the compressor system," because that ammonia would go right through and neutralize some of my acid, and make me think I had ethylene oxide when I really did not have it. That was really why we put it in. And I and my assistant talked it over and said, "Well, it won't do any harm, and the patent says it will help, let us put it in." We need to have something to take out the ammonia; and the air is moist anyway.

Q. (By Mr. Johnson) I neglected to ask you, Dr. Lamb, this question. You frequently used the word "we" and now you refer to an assistant. Who was he, and tell us whether he had any previous knowledge of the reissue, or of the patent, or what was to be done?

The Witness: I had a very satisfactory assistant named Mr. Damon, Mr. Edwin Damon, who is an excellent analyst and operator, but who had never had any experience with catalytic studies, nor indeed with methods of testing catalysts, but he was an excellent analyst. He, for instance, had never seen or used a flow meter before. He constructed them and calibrated them with the utmost care and skill. He was an excellent analyst, and actually performed the analyses that I referred to, although I verified substantially every end point that he took, and the weighings that he made.

Q. Well, did he know anything about this situation before? A. No, he knew nothing except what I told him about the problem that had been presented to me.

The Court: Now, I have one other question. I realize this may be anticipatory, but if we go ahead with the testimony and at the same time get at the point of law, so much the better. If I understand you, Prof. Lamb, it would follow as a necessary result of your statements that you do not think there is any real materiality in what is stated in the French patent, No. 771,650, by the French Societe, to the effect that an improvement had been found. It is expressed in this language——

Mr. Johnson: I believe the witness, your Honor, has never read that patent.

The Court: I am assuming he has never read it.

The Witness: No.

The Court: But you heard the testimony about it?

The Witness: I heard some testimony.

The Court: The statement is made, the applicant has improved this process—well, I might read back a little: "The present invention relates to the preparation of ethylene oxide by the direct oxidation of ethylene in contact with pure oxygen or air, and the reaction or direct oxidation requires special precautions against explosion hazards. In connection with the industrial oxidation of ethylene it was suggested as early as 1904 that water vapor be introduced into the reaction. The explosion hazards are thus caused to disappear. But the oxidation products have always been aldehydes. The applicant has improved this process by bringing into play catalysts suitable for obtaining a specific reaction which gives ethylene oxide as the oxidation product. The thus improved process marks a considerable step forward in the technique of the production of ethylene oxide by the direct oxidation of ethylene. But the necessity of vaporizing large amounts of water requires an expenditure of calories, which obviously contributes its share to the production cost of the ethylene oxide obtained."

Now, I understand in that latter sentence he is talking about the patent that you used. "The present invention"—he is talking now about his alleged improvement—"remedies this drawback. It has as its object a process for preparing ethylene oxide by the direct oxidation of ethylene without water vapor and with no explosion hazards. This process is characterized principally in that the ethylene-oxygen or ethylene-air mixture is diluted with a large amount of an inert gas."

Now, if I understand you correctly, you say that those are, for all practical purposes, from your scientific viewpoint, those are merely alternate things, and that both are equally suggested by the original patent.

The Witness: Well, the answer, I think, is "Yes." Certainly, as far as my general knowledge is concerned, and my own experiments are concerned, the cost of vaporizing the water in that wash bottle of mine was certainly negligible—it did not cost anything—and there was moisture in the air supplied gratis, and it cost nothing. I can not see the novel contribution. Of course, it is known that with an exothermic reaction you can not have too high a concentration of your combustible materials, and you must provide for the dissipation of the heat; and a great variety of diluents could have been used, involving no exercise of invention, I should think.

The Court: Then, you disagree with the emphasis that Professor Taylor and Professor Reyerson put on this use of water?

The Witness: Certainly, I do, as far as anything I know.

The Court: Go ahead, now.

CROSS EXAMINATION.

By Mr. Barrows:

Q. What was the sort of compressed air you used? A. Laboratory compressed air. The compressor is not actually in our laboratory, but it is in an outside university building, and it is piped through the ground to our laboratory, and comes to a fairly good-sized storage tank, with a pressure regulator in our sub-basement, and is then piped from our storage tank to various parts of the laboratory.

Q. What pressure is that air held at? A. The air in the lines, the distributing lines, about 4 pounds above atmosphere.

Q. And is that the highest pressure that it is compressed to in the compressor compressing it? A. Well, I can not tell you about the pressure in the University compressor line, but I do not think it is a very high pressure line.

Q. Well, would that pressure remove part of the normal moisture content of the air and reduce it somewhat? A. Well, it would depend somewhat on the prevailing

humidity. But if the air were 100 per cent. humid that entered it, it would precipitate moisture.

Q. Now, you said you were asked on April 27th, as I recall it, to carry out the experiments; is that correct? A. That is correct.

Q. And the first experiment recorded in your report is May 26th. Is that the first experiment you did? A. Yes, it is.

Q. It was about a month after you were asked to do it before you carried out the experiment? A. Yes, that is right, before we made the run to test the catalyst.

Q. How long a time was required in making the catalyst? A. It wasn't very long (referring to records), two or three days is my recollection. We heated the material overnight at one stage of the operation and then it is also true that after we got our silver oxide we made a number of small scale heating experiments to see how it behaved at different temperatures. We found that, placed in a little glass tube and stuck right into the flame, it heated up and almost exploded, so sudden was the decomposition.

Q. With the air? A. Yes. And then we heated some more gradually, and so on, and tested it with a little hydrogen and found it difficult to control.

Q. Hydrogen and air? A. No, first we tried out straight hydrogen. Of course, that was very difficult, and heated slightly the temperature rose with great rapidity.

At that time I had read pretty carefully an article by Professor Lewis, who was my old teacher, on decomposition of silver oxide, and I then proceeded to decompose it pretty much according to his directions. He gave the temperatures needed for rapid decomposition of silver oxide, and from that point on the preparations were pretty rapid so that I should say it took us perhaps three days all told in making this catalyst.

Q. Where was that article published, or book published, you referred to? A. Well, I think it was published in two places, but my particular copy was the article that he published in the proceedings of the American Academy.

Q. What date, do you know? A. It was about 1905 or six.

Q. These preliminary tests of taking silver oxide and heating it under various conditions were all made before you finally made the catalyst, is that correct? A. Yes, that preceded it by perhaps a day before we fixed on our heating schedule.

Q. Then this experiment on page 5 of silver on the carrier dated June 3, 1940, was in between the dates of the June 1 and June 9 experiments recorded on page 4?

The Court: What has all this got to do with our little problem? What is the necessity of taking all of these dates and so forth?

Mr. Barrows: . . .

I want to ask him if this record of June 3 was reported to the attorneys for the plaintiff before he carried on his experiment of last Sunday, June 9, just three or four days ago.

A. (After referring to records) Yes, I think I told the attorneys about this experiment when I came down here last week.

Q. Were you asked to make another experiment last Sunday? A. No, I was not asked. As a matter of fact, it is a little matter of professional pride. I felt that we ought to have collected the CO_2; I knew how to do it, and I had hesitated to take the trouble, we really were terribly busy at this particular time, I had a thousand other things to do besides these experiments at this time of the year, and I just had time to get those U tubes fixed up and tested and carry through them a quantitative collection of CO_2.

Q. You have had a great deal of experience in carrying out catalytic experiments, have you not? A. Yes, in the old days I had a good deal of experience.

Q. And you have had a good deal of experience as an expert in patent litigation? A. (After a pause) I can not — I suppose it is dull of me, but I can not seem to remember testifying in a patent case on catalysts before. I suppose not.

Q. But you have had a good deal of experience in various connections with catalytic work? A. Oh, yes, indeed.

Q. And you are a member of the Committee on Catalysts and have been for a number of years? A. I don't think so recently, but I was originally on that committee.

Q. And you have made it your business to keep track of the development of the literature on catalysts? A. I can not say I do. I am sorry, but I do not.

Q. In former years that was your custom? A. It was during the period up until 1925 or 1926; I was very much interested then in that field and I had time to do it, and I did keep pretty good track of catalytic work at that time.

Q. Of course, this work which you did and which you recorded was done at the request and at the expense of the plaintiff here? A. Yes.

Mr. Barrows: That is all.

REDIRECT EXAMINATION.

By Mr. Johnson:

Q. You said that you made this apparatus or this assistant of yours made it? A. Yes, Mr. Damon actually built a good deal of this, and some of the more difficult features, the flow meters, were manufactured by the glass blower in our own glass blowing shop.

Q. And then those were calibrated? A. Yes, it took weeks to get that analytical apparatus effected.

Q. Does that account for the time lapse between April 27, when we asked you— A. Yes, the first three weeks were spent almost exclusively in getting ready the analytical apparatus and in familiarizing ourselves somewhat with ethylene oxide, because I did not know much about ethylene oxide, I had never worked with it before, so we got a cylinder of it and fussed around with it and became fairly familiar with it.

Mr. Johnson: That's all.

The Court: If you did nothing more, if you stopped right here and did nothing more in the way of experimentation or laboratory work, and you were asked by some third party industrially concerned with the thing,

whether from what you had done, under the teachings of this patent, you were satisfied that that company could produce with commercial success, that is, profitably from the point of view of conversion, ethylene oxide, you would answer that, yes? A. Yes. That is a surprising thing, too, your Honor, because when I first saw this patent I was more or less dumbfounded, to tell you the truth, to think that you could realize this reaction, which, to a chemist, is quite a surprising reaction, and I did not know that it had been achieved.

The Court: Do you mean that you sympathize with a Court that has to sit through it?

The Witness: Yes, I do. I should certainly tell anybody that asked me that it looked to me, from my limited experience, it looked easy.

The Court: You mean now you would tell them?

A. I would have told them before I came into this court. I mean, after these experiments had been performed.

The Court: In other words, if you were asked for such advice as I have just suggested, you would be willing, on the experiments that you have made and none other, and the study that you have made and none other, to give them an affirmative answer, yes?

A. Absolutely.

The Court: That's all * * * [770-795].

* * * * * * * * *

Thereupon——

DR. ARTHUR B. LAMB, resumed the stand and testified further as follows:

Direct Examination.

By the Court:

Q. My main question relates to this: Have you, since you testified, had a chance to study the reissue patent? A. Last evening I read it for the first time, so I have some familiarity with it.

Q. Well, then, do you feel you are in a position to testify more fully now from a scientific standpoint as to whether you find any material difference on this question that has been dwelt upon so much, the alleged departure, in that water is required in the first patent and not in the second, as an addition? A. I certainly have not heard anything that has changed my ideas about water as an ingredient in this reaction. I was a little disturbed at Professor Taylor's failure to give all of my reasons for putting the water in. In my experiments I needed to have an indicator for ammonia; and I discussed whether or not to put the water bottle in, that would add additional water. And, as I have stated in my testimony, I thought that after all water is recognized in this patent as advantageous, and it would certainly do no harm if you put it in. It might even get me larger yields of ethylene oxide. So there were really two reasons why it went in, although the use of it as an indicator was the primary one. But I certainly was astonished when Professor Taylor said that I, who have some familiarity at least with catalysis, and had endeavored through my best efforts to follow the first patent, had inadvertently apparently been doing the second patent. It does not seem to me that they can differ very greatly if a person skilled in the art practices the second one when he thinks he is practicing the first.

Q. Well, now, that leads to the other question I was going to ask you. You practiced the first, and have given us the benefit of the results of that practice? A. Yes.

Q. Now, you have since read the reissue? A. That is true.

Q. And you had not up to that time read the reissue? A. That is true.

Q. And you state since you have studied it? A. I have studied it.

Q. Well, now, suppose in the light of everything you know on both patents to date, that is, the experiment you made under the first, and your study of the second in the light of the first, suppose you were now given this second, or reissue, patent, and asked to return to your laboratory and follow out the teachings of the second, would you or would you not feel you were required to make any de-

parture from what you previously did under the first patent, and if so, in what respects? A. I do not think I would have done a bit differently.

Q. I am referring now to claims 8 and 9, because the other claims are identical. A. Yes, Claims 8 and 9 mention the carrying on of the reaction in the presence of water. And I think I would have done just the same, only it would have been a little easier for me, because I would not have had quite so much hesitation in regard to the selection of the catalyst. The number of optional catalysts is somewhat reduced in the reissue patent. It would have been a little easier for me. But I think I would have done the same experiment.

Of course, this matter of water, too, I think is a little hard to see in its correct light, on account of the numbers that are involved. We speak of 1½ per cent. of water as very little water. Well, it is, compared to the total bulk of air, or the total amount of ethylene that went in, or the total amount of materials that flow through the system. But compared to the amount of ethylene that you are using, particularly compared to the amount of ethylene oxide you get, your water content is large. The amount of water that you get in the effluent gases, under the conditions I worked, and they were pretty nearly comparable to industrial conditions, was greater than the ethylene oxide then produced. In other words, although the percentages are small, the relative number, the relative amount of water vapor in these reacting gases is very large, even as I carried it out * * * [1174-1178].

* * * Cross Examination.

By Mr. Barrows:

Q. Dr. Lamb, I believe you said you found it easier to read and follow the reissue than the original. Just what did you mean by that? A. Well, may I have a copy of the reissue patent?

(Handed to the witness.)

The Witness (continuing): In the first patent there is a long and very numerous list of catalyzers that may be

used—I think there has been discussion about that in the testimony—while in the reissue the statement is relatively brief about the catalysts that may be used.

"The surface catalyst is preferably silver, as stated, but other metals, such as bismuth and antimony may be used. Likewise, oxides of these metals may be used. Experiments have also indicated that the catalyst may be silver activated by the addition of small amounts of gold or copper or iron."

I do not seem to see anything else in the specification about the chemical nature of the catalyst.

Mr. Watson: I call your attention to line 47.

The Witness: Well, I do not see any additional catalysts mentioned there, except that it speaks of silver in finely divided form.

Q. (By Mr. Barrows) Silver in finely divided form imports such a catalyst as you prepared from the oxide, by reduction? A. Yes.

Q. Now, you find no mention of silver in finely divided form in the original patent, do you, except in the reference to a silver colloid in the modus operandi 3, and reference to silver in powdered form in modus operandi 2? A. No, I think there is no other mention in the specifications * * * [1179-1180].

* * * But, of course, I could point out that I did not need much instruction on the importance of using silver in a finely divided form.

Q. Well, if you had followed Example 3 of the original patent you would have used silver in the particular form of a silver colloid, would you not? A. Yes.

Q. And the catalysts that you prepared and used would not have been suitable for use in Example 3 at all? A. I don't know about that, Mr. Barrows.

Q. At least the catalyst you used was not a silver colloid? A. It was not in suspension.

Q. Can you say whether or not it was or not a silver colloid? A. I can't say categorically because of the fact I don't know the number or the size of these silver par-

ticles that were the constituent particles of my silver pellets.

Q. But you do find in modus operandi 2 reference to silver in powdered form? A. Yes.

Q. And also in the form of wire gauze? A. Yes.

Q. And in the example you also find the use of a mixture of ethylene, air, water vapor and hydrogen, do you not? A. Yes.

Q. And you made no attempt to carry out the process of that example at all, did you? A. I did not, in so far as the inclusion of any hydrogen was concerned. I did not add any hydrogen to the mixture. As I remember it, hydrogen was spoken of as a particularly optional addition in some place in the patent.

Mr. Fairbank: Line 4?

The Witness: Line 5, it is * * * [1181-1182].

* * * * * * *

TESTIMONY OF GEORGE H. LAW

Thereupon——

DR. GEORGE H. LAW, a witness of lawful age, produced on behalf of the plaintiff in rebuttal, having been first duly sworn, was examined and testified as follows:

Direct Examination.

By Mr. Johnson:

Q. Will you state your name, age, residence and occupation? A. My name is George H. Law; age, 35; I reside at South Charleston, West Virginia; chemist is my profession.

Q. And you work for the plaintiff company, is that correct? A. Yes.

Q. How long have you been with the company? A. Approximately eleven years.

Q. And you, in the early part of 1936, carried out some tests on the original Lefort patent? A. Yes, I did.

Q. What were your qualifications at that time? A. You mean my scholastic qualifications?

Mr. Johnson: Yes.

A. I graduated from Lehigh University in 1929 with the degree—excuse me, in 1926, with a degree of chemical engineer; from Yale University in 1929 with a Ph.D. degree, majoring in organic chemistry. I believe that approximately half of my time while I have been employed by Carbide and Carbon Chemicals Corporation has been devoted to working on vapor phase contact catalysts • • • [796-797].

• • • The Court: I think for the present anyway I would like to limit this witness' testimony to answer to such questions as follows:

Q. Did you follow the teaching of this original U. S. Patent in making the certain experiments for your company? A. Yes, sir.

Q. When did you do that? A. February, 1936.

Q. Why were you asked to do it? A. I am not quite sure.

Q. You weren't given any reason except to go ahead and do it. A. By my immediate superior.

Q. How many experiments did you make? A. Over a period of time?

Q. In all, that have been submitted here.

Mr. Watson: That is up to the time we got the rights.

Mr. Johnson: Prior to the time you purchased the patent.

The Witness: I am not quite certain when we purchased the patent.

The Court: You made numerous ones.

The Witness: I would say that book contains about twenty experiments.

By the Court:

Q. And you have heard the testimony of all the other experts here? A. Yes.

Q. And you have heard the testimony of the two experts for the other side, who testified with respect to the apparatus they used, and their claims that they made ethylene oxide successfully? A. Yes.

Q. You heard that? A. Yes.

Q. Did you follow substantially the same methods in taking the teachings of the patent? A. I think my methods were essentially those of Doctor Lamb, except that we extended that work to supports.

Q. You did what? A. We extended that work to include supports, catalyst carriers.

Q. What results did you achieve? I mean generally speaking, out of a number of experiments that you performed, what was the conversion percentage of ethylene oxide that you got? A. After about four, or possibly five weeks work here on rather crude laboratory apparatus, I believe our maximum yield was about ten per cent. and our efficiency about fifty per cent. We were using at that time a silver catalyst as I recall, at least part of the time a silver catalyst which was supported on the filtros that has been discussed.

Q. Do you claim that was satisfactory or not from an industrial point of view? A. Well, it would be rather hard to say. As I said before, we were operating on a laboratory scale and on relatively crude apparatus and that indicated to us, I think all the members of our company, that there would be no question at all but we could go ahead from there and operate an industrial process.

Q. Were all of these experiments you referred to borne out—I mean represented by the figures that have been put in evidence? A. That figure I gave you we obtained toward the end of these experiments. In the first two experiments we did not obtain any quantitative data and as we progressed we got more and more quantitative data and toward the end of this series of experiments we actually got quantitative data that had the values I have told you.

Q. What did you say to your company? Did you have to make a report to your company? A. No, sir, as soon

as we got the first qualitative test, Doctor Curme came down the next week, he is our vice president in charge of research, came down the next week and insisted that we actually isolate some of the material. You understand, the original tests were obtained by chemical methods, not by isolation, and I gathered he was very skeptical whether we actually made any and he came down and we demonstrated that we could isolate some ethylene oxide and I think immediately following that, even before we had those figures I reported to you, he was satisfied we could go ahead from there. That is my impression.

Q. And that ended your work? A. Oh, no, sir, I went ahead then, we had a great deal of engineering experimental work.

Q. I mean it ended your work on these experiments, did it? A. We went ahead then and did large scale work. There are a great many things to explore in engineering work and we went ahead from there largely, I would say, on development work.

Q. What do your notebooks cover? That is one thing I want to get, that are here in evidence?

Mr. Johnson: They are not in evidence, your Honor. I inadvertently misled you. They were given to defendant's counsel in connection with the deposition of Doctor Curme which took place prior to trial and it is not in evidence.

By the Court:

Q. What do your notebooks cover? A. That notebook covers a period I think of approximately five weeks or longer.

Mr. Johnson: February 11th, 1936 to March 6th, 1936.

The Court: Were you actually producing ethylene oxide during that period? A. On a very small scale on the apparatus we then had.

Q. You admit it was on a smaller scale than you later produced and on a smaller scale than Doctor Lamb produced? A. No, I think if anything a little larger than

Professor Lamb, in that same order of magnitude. I believe we started on a somewhat larger scale and possibly five or six months later developed——.

Q. I mean when I said on a large scale, I am speaking of terms of conversion or efficiency? A. Excuse me. Yes, it was smaller than Doctor Lamb.

Q. You did not get as good results in any of the experiments covered by that notebook as he stated on the stand he got? A. No, sir, we didn't.

Mr. Johnson: We offer this notebook in evidence.

Mr. Edmonds: Is that to be received in evidence? If so, we will have to go into these tests in some little detail.

The Court: Why do you have to go into these tests? I don't want to hear them. Here is a witness who admits, as I understand it, that up to the time covered by this book, by this process he wasn't drawing ethylene oxide off in satisfactory quantities but that he did it later. Isn't that in effect what you said?

The Witness: By satisfactory——

The Court: What you call commercial purposes, you were not satisfied that you were getting enough conversion?

The Witness: We were not as satisfied with what we got then as with what we did get later.

(First notebook of witness Law marked "Plaintiff's Exhibit 14.")

By the Court:

Q. And later on you say you did find out and produced an efficiency of conversion, increased it, I mean? A. Yes.

The Court: What on earth is the sense in going through all the figures? After you have done it, the witness admits the very thing that presumably you want him to admit.

Mr. Edmonds: If he will admit further that the first half a dozen efforts were fruitless, that is ended there.

By the Court:

Q. Were they fruitless or not? A. We did not obtain any ethylene oxide in the first series of tests referred to, requiring about twenty-four hours of actual operation.

Q. They were in effect failures? A. We did not get any ethylene oxide.

Mr. Barrows: And in those tests you used silver gauze catalyst.

Mr. Johnson: Your Honor will permit me to ask the same question Mr. Barrows wanted to ask because that is what I want explained in the record.

The Court: Very well.

By Mr. Johnson:

Q. Why did you use this silver gauze when you first began your work? A. We wanted first of all, if I recall, to employ silver without anything else to give us a convenient thing to use to insure the formation of a form of pure silver. We used it knowing that it is a massive smooth form of silver which would ordinarily not be considered to have catalytic activity, and we also knew at that time that metals of that type, smooth sometimes, acquired a catalytic surface in operation. It was known that is the case in the platinum gauze used in ammonia oxidation and also occurs in silver gauze used for the oxidation of hydrogen, as I recall. We selected it and ran it knowing those things. As I say, the first test with the gauzes lasted about fifteen hours and we thought perhaps it had been contaminated and put another in and ran that about eight hours.

Q. Was that method enough to develop the surface you are speaking of? A. No, our gas analyses indicated that it had no catalytic activity in the range over which we wished to operate; that is, below 400 degrees, and that apparently this surface had not as yet developed. We were getting no oxidation of ethylene to speak of below 400 degrees.

Q. May I ask the witness one more question? What he did to develop that surface on the silver gauze? A. We dipped the silver gauze in molten silver nitrate again

with the idea of using silver alone and decomposed it by heating it thermally, as I recall, about 600 degrees, and we did get ethylene oxide with that catalyst.

Q. This further work you speak of after you acquired the patent, was that engineering or research work like the previous? A. Primarily it was engineering work. In the course of that we were able to develop some improvements which I would consider more in the light of research work. I don't mean to say the process would not work without these things because it will, but we were able to develop some improvements. Primarily what we did was engineering * * * [802-812].

* * * * * *

TESTIMONY OF EMORY P. BOYNTON

* * * The Court: How many more witnesses have you?

Mr. Watson: I think I have to call Mr. Boynton, who was called and put at the disposal of Mr. Edmonds this morning.

The Court: What do you want to ask him now?

Mr. Watson: I wish to ask him if he had in front of him when he prepared the application for the Reissue patent in suit the late French or British patents of the Societe. It has been intimated by the defense that the Reissue was prepared with the knowledge of those late French ones and with the deliberate motive of reaching out to grasp something not in the original.

The Court: I thought that was asked him or certainly impliedly asked him this morning. That was implied in his answer when he said that he did not know of anything, that he was speaking directly in connection with an application akin to the British application, wasn't it?

Mr. Watson: He was saying that he made a diligent search of all the United States applications ever filed by this French company at the time the plaintiff bought and that he found no record.

The Court: Yes, of an American application.

Mr. Watson: Yes, of an American application.

The Court: And now you want to ask whether he had the third French patent before him when he did what?

Mr. Watson: When he prepared the application of the Reissue of the original.

Thereupon——

EMORY P. BOYNTON, a witness of lawful age, produced on behalf of the plaintiff in rebuttal, having been first duly sworn, was examined and testified as follows:

By the Court:

Q. Do you understand the question? A. Sure.

Q. What is your answer? A. Your Honor, I had nothing before me except the original patent. I had not heard at that time of British specification 431 something or other and the French patent 771,650, I believe it is.

Direct Examination.

By Mr. Watson:

Q. Do you know whether you had those before you at the time you obtained an allowance of the Reissue? A. No, sir, I did not see either of those documents or any copies of them or never heard of them until after the Reissue patent had been issued.

By the Court:

Q. Well, in a few words, what were you motivated by primarily, what were you acting on? A. On the strength of my investigation of the original patent I had decided that it was a very fit and proper case for the application of the Reissue law. It seemed to me that here was a man who had, through what I would call gross ineptitude of a patent solicitor, been given claims of extremely narrow scope for an invention which is a basic invention if I ever saw one and to me it seemed that was a miscarriage of justice that in all equity should be remedied and that is exactly what our Reissue statute is for. Having decided that and having before me the original patent, of course, I had at least six out of the seven French disclosures, that is the 729 something or other patent.

and its additions and the 739 French patent, and I think its addition, I had at least six of those before me, which were the documents referred to in the oath of the original patent. I believe one of them was missing at the time I drew the application for the Reissue but I had at least six of them and having looked at six and the original patent I then wrote the first of the Reissue specifications and that was not written in September, '36 as has been said here. It was filed then but it was written sometime earlier in that summer and forwarded to France to be executed by the inventor, Mr. Lefort, but he was on his holidays and was then, I believe, no longer connected with the French company and it was not until late, not late, but early in September, about the middle, perhaps, the 10th, I think that he was located and brought to Paris where he signed the Reissue application and then it came back and it was filed in September.

Q. You mean the events or the series of events that more directly than anything else led you to prepare the application was the series of events covered by the original Lefort patent, is that it? A. That is right, but excluding this one that has been called the third French patent.

Q. You mean that was the main thing that started you off? A. Yes, I studied these patents and the original patent and its history in the Patent office and from that I concluded that Reissue should be possible and that this was a very proper case for the Reissue statute.

Q. Under whose direction were you doing that? Or was that your idea? A. I am employed by the plaintiff corporation as a patent attorney.

Q. What first started you off on it? A. I don't recall just when I first heard of this Lefort patent but at about the time of these negotiations, at the time they were being conducted by cable between our New York office and our representatives in France to acquire rights under the patents, I was asked to say whether or not that patent would cover the process disclosed in it and I found a wealth of disclosure and great poverty of claim, which on investigating the facts in the file history and in these French patents struck me as being entirely inequitable.

Q. Let's see, I am a little confused. Are you talking as of what time? There is a period here when they were believed not to be any good, the French patents, by that company? A. Oh, no, this was after Doctor Law had gotten positive results.

Q. This was all after he had gotten positive results? A. Yes, that is correct. I am speaking of the time probably in May, I believe, April or May of 1936, which was after Doctor Law had gotten his results and Doctor Curme and his associates had completed their negotiations to acquire rights under the patent.

Q. That is under the American patent? A. Yes, under the American patent. We have no rights under any other patent.

By Mr. Watson:

Q. Mr. Boynton, a photostatic copy of the assignment of the patent in suit has been offered in evidence by the defendant. Is that the common form of assignment in use by your company, do you know? A. Well, it was not a printed form, you know, Mr. Watson. The assignment was drafted specially for this instance to cover the assignment to us of the original United States Patent and the only United States application that we have ever heard about and in it we put the usual provisions that we always put in all of our assignments for divisions, or extensions and continuations and reissues. At the time that assignment was drawn I had already recommended and had started the work of preparing the Reissue application, so I imagine I considered these questions with more than usual attention, but we put the matter in all of our assignments, those that are used within our own company for transferring patents or applications from individuals to the company, from one unit of a corporation to another unit and from outsiders and in every case we provide for the Reissue rights to be included and the signing of the oaths and that kind of thing. As a matter of fact, this is standard among all corporations and I happen to know it is a usual practice of the defendant in this suit.

Q. How do you know that? A. Because at one time or another I acquired a certified copy of an assignment

of a patent that has been discussed here, the Carter patent, 2,125,333, and it is a printed form used apparently, rather generally. This is an assignment from Ray M. Carter to the Air Reduction Company, Incorporated, and on a printed form with respect to Reissues and that sort of thing that was put in the assignment from the French company to the Carbide and Carbon Chemicals Corporation of the original U. S. Lefort Patent * * * [898-905].

* * * DIRECT EXAMINATION (continued).

By Mr. Watson:

Q. Mr. Boynton, have you made inquiries from the United States Weather Bureau in Baltimore with respect to the humidity in the air over a year's period? A. Yes, sir, I obtained from the Weather Bureau, the Customs House here in Baltimore, a printed form 1030, which contains data on the relative humidity in temperatures and other meteorological data for a period of the past approximately thirteen months. I had that calculated by one of the engineers of our company, to show the per cent. by volume in the air for the various days, of water vapor.

Q. Is that the tabulation on the sheet which I hand you (handing paper to witness)? A. Yes, this is the tabulation.

Mr. Watson: I offer that tabulation sheet of the Baltimore weather conditions on water vapor in the air, as Plaintiff's Exhibit 19.

(Tabulations of Baltimore weather conditions on water vapor in the air marked "Plaintiff's Exhibit No. 19.")

Mr. Watson: That is all.

CROSS EXAMINATION.

By Mr. Barrows:

Q. Mr. Boynton, when were you admitted to the Bar? A. I don't remember the month. It was either October or November, 1933.

Q. And in 1935 and '36 you were employed by the Carbide & Carbon Chemicals Corporation, in the Patent Department. A. That is correct.

Q. Who first called your attention to this original Lefort Patent? A. I believe that it was Dr. Perkins of our South Charleston Laboratories, who asked me to get some copies of it for him.

Q. And who asked you to investigate this patent? A. One of my superiors. I don't recall whether it was Mr. Scheffler, who is the head of our Patent Department or Dr. Curme.

Q. And you did make an investigation and report on this original Lefort Patent? A. Yes, I made an investigation.

Q. And you knew at that time what the process of the plaintiff was, as developed up to that time? A. No, I cannot say that I did. I probably knew that they were working on the direct oxidation of ethylene to ethylene oxide with the silver catalyst, but I had no idea of the procedural details.

Q. So your report was not then an infringement report at that time? A. Well, it was a rather comprehensive report on the subject of the patent, as respects its possible merit and scope.

Q. Showing that the patent as it was developed was limited to the use of water. A. I don't recall that I drew any such conclusion, Mr. Barrows, although having seen the claim so frequently lately I would say that water or steam was included in all of the seven claims of the original patent.

Q. Now, then, the idea of reissuing this patent was your idea, I take it? A. Well, in my report I pointed out the facts that I spoke of yesterday, to the effect that the invention seemed to be far broader than the claims, and the file history showed that the patent attorney who had prosecuted it in this country had done a very poor job of getting claims even partly commensurate with the scope of the invention, so I said that it seemed to me to be a case where reissue should be possible, to improve the protection for the invention that was disclosed.

Q. And you were the one that prepared the reissue application? A. That is correct.

Q. And the reissue oath? A. Well, I undoubtedly had assistance in preparing both of those things, but I prepared them, yes.

Q. You prepared the assignment of this original patent from the French company to the plaintiff? A. No, I formulated what should go into the assignment, but the actual assignment was drafted by another attorney for the Corporation.

Q. Do you recall that in that assignment there was a pending application included as well as the issued Lefort Patent? A. That is correct.

Q. And did you take over the prosecution of the application as well as the handling of the Reissue? A. That is correct. Some time, I believe in November of 1936, I finally got the file from the United States Attorney who had previously handled it, of that Francon application that is named in the assignment, together with the original patent.

Q. Did you prepare or aid in preparing the Curme affidavit that was filed with the Reissue application? A. Yes. As a matter of fact, I attended to the actual typing and the legal portions at the beginning and end of that, but I had very little to do with the substance of that, Mr. Barrows.

Q. Did you go down to Charleston, to Dr. Curme, when he prepared his draft of it? A. Yes, I was in Charleston at that time.

Q. And you went over with him the records of the work that he said that he had considered at that time? A. No, that was provided to him by the men in the laboratories. I was more concerned in my duties with him in Charleston at that time in discussing the prior art, which he discusses in that affidavit.

Q. Mr. Boynton, I show you Defendant's Exhibit 4, which is the file wrapper of the Reissue application (handing exhibit to witness). I call your attention to page 2 of the Specification of the application as filed, and ask you if the statement of the object of the invention which is given there is your statement of the object, one which you prepared? A. Yes, I wrote that.

Q. On page 1 of this reissue application, Mr. Boynton, you will note at the bottom of the page a reference to silver powder or silver wire gauze as examples of the preferred catalyst? A. You mean on page 2, don't you?

Mr. Barrows: Page 2 near the bottom.

A. Yes.

Q. And do you recall that in the original Lefort patent silver powder and silver gauze was also mentioned in Example 2? A. That is correct.

Q. Now, I show you the file wrapper of the Francon application, which I understood you to say was the one that was assigned at the same time as the Lefort patent, and of which you took over the prosecution, is that correct? A. That is correct.

Q. And the amendment which I show you and which was filed in the Patent Office on November 28, 1936, and which is signed with the name of Mr. Greenwald, is one that you prepared? A. May I see it, please? (Paper handed witness by Mr. Barrows.) Yes, Mr. Greenwald is the head of the patent department, and he with Mr. Sheffler, our general patent counsel, hold the power of attorney in probably 90 per cent. of the cases we prosecute, but I prepared that * * * [926-934].

* * * Q. Mr. Boynton, in this amendment I have referred to, and in discussing the original Lefort patent, you said that the Lefort patent mentioned two physical forms in which the catalytic material may be used, one of these is, "wire gauze" and the other is "powder form, as mentioned at lines 28 and 29, page 2, column 1, of the patent." And, further on in that same paragraph you said, "The term 'powder' almost without exception and invariably as applied to metals connotes something which has been ground, or in which the particles are more or less spheroidal."

That is the representation which you made to the Patent Office in this amendment, is it not, under date of November 28, 1936? A. I prepared this amendment.

Q. And you find there the statements I have quoted? A. Yes, I find them.

Q. How old are you, Mr. Boynton? A. Thirty-one.

Mr. Barrows: That's all.

REDIRECT EXAMINATION.

By Mr. Watson:

Q. How long have you been in the patent department of the plaintiff company? A. Ten years.

Q. Was the subject of this Francon application a special physical form of silver lamina torn in sheets of extreme thinness? A. Yes, it had to do with the proposition of beating the metal foil, silver foil, into extremely thin sheets and tearing it into very small pieces so as to form thin small flat particles.

Q. When you prepared and prosecuted the reissue patent in suit did you exclude the use of silver gauze? A. Oh, no. I thought that the term "finely divided" said in one phrase practically everything that had been said about silver, which was said a great many times throughout the original Lefort application—or, rather, the original Lefort patent, and which includes—well, it would include a silver gauze or any other physically prepared form of silver which is fine, like ground up metal, powder, and chemically precipitated silver, such as that silver colloid which was in the third example, being formed by chemical precipitation and it would include chemically prepared silver in various forms, because when he said that he would use silver on a support, it is very difficult to imagine hammered out or ground up silver being so applied.

Q. Do you know whether your company has ever used the particular silver catalyst of that Francon application? A. I am sure we have not.

Mr. Watson: That is all.

The Court: Where did you study? Did you go to any engineering school first?

A. Yes, I did.

The Court: Where?

A. I graduated from Armour Institute of Technology as a chemical engineer.

The Court: And then where did you take your law?

A. New Jersey Law School, Newark, New Jersey.

The Court: That's all.

Mr. Barrows: One more question: Mr. Boynton, you know about these experiments that were carried out early in 1936 using first a silver gauze in which no conversion or formation of ethylene oxide is obtained, and then using silver gauze as a carrier or support by depositing an extra surface on that, you are familiar with those experiments, are you not?

A. Yes, I first saw the results of those at the same time you did, Mr. Barrows, or perhaps a day before you saw them.

Mr. Barrows: That's all.

• • • [936-939].

• • • • • •

Thereupon——

EMORY P. BOYNTON, a witness, recalled by the Court, testified further as follows:

By the Court:

Q. Will you state just briefly, Mr. Boynton, what you thought the Reissue patent or the original patent ought to claim which it did not claim and which you sought to put into the Reissue claim? A. I thought that Mr. Lefort had made an invention which probably should have entitled him to far broader claims than were ever advanced in either of his applications and at one time, when I first wrote the Reissue application and first filed it, I had in mind trying to get that invention secured for him because he obviously had done something that had been—everyone had been near it but no one had actually hit upon this direct oxidation of ethylene to ethylene oxide. It was a totally new sort of chemical reaction. On studying the matter further and particularly after the first Reissue application was filed, it was apparent that because of the way that the French solicitor

had prepared the original U. S. Application from these French patents that it would be virtually impossible to claim the invention broadly so as to cover it whether or not water was even present, added or introduced or formed and with any catalyst and in the scope that an absolutely basic invention should be entitled to be protected. So it was plain then that because of the nature of the patent handling rather than his invention that it would have to be an invention which was clearly within the original disclosure and within his original statement of invention and that was to cover the oxidation of ethylene to form ethylene oxide directly by oxidation with molecular oxygen in the presence of a catalyst and in the presence of water and the catalyst had to be one of those that was named by him and it was perfectly obvious from the original specification that silver or silver modified slightly or something closely akin to silver would have to be the catalyst.

Q. As a matter of fact, you narrowed the catalyst situation in the Reissue, didn't you? A. Yes, it was the intention to do that.

Q. You did not broaden the situation as to the catalyst, you rather narrowed it? A. Yes, he had no real foundation so I found for including certain of these catalysts that were named in his specification and rather than have in there a possible confusion to anyone looking at the specification, it was a case of narrowing down that disclosure of the catalyst and covering them in the claims commensurate with that revised description of the catalyst.

Q. In working this up, did you have the benefit of the help or advice of any technicians other than those who have been here to testify? A. I did not even have the benefit of the help of all of those because Professor Stevenson and Professor Lamb, who testified for the plaintiff here, were not associated with us at that time, but Doctor Curme and Doctor Law and Doctor Perkins were available and I had, of course, consulted with Mr. Watson's firm, Mr. Watson and Mr. Johnson, in that connection * * * [1003-1006].

Reissued May 18, 1937 Re. 20,370

UNITED STATES PATENT OFFICE

20,370

PROCESS FOR THE PRODUCTION OF ETHYLENE OXIDE

Theodore Emile Lefort, Paris, France, assignor, by mesne assignments, to Carbide and Carbon Chemicals Corporation, New York, N. Y., a corporation of New York

No Drawing. Original No. 1,998,878, dated April 23, 1935, Serial No. 699,378, March 22, 1933. Application for reissue September 25, 1936, Serial No. 102,613. In France March 27, 1931

9 Claims. (Cl. 260—54)

This invention provides a specific and novel process for making ethylene oxide. It essentially consists in causing ethylene to combine directly with molecular oxygen at temperatures of about 150° to about 400° C. in the presence of a surface catalyst which favors the oxidation of ethylene to ethylene oxide under these conditions. The process can be carried out at atmospheric pressure, and it may also be conducted at pressures above or below atmospheric pressure.

This process is characterised by an entirely unique reaction in which the direct addition of a single oxygen atom to the ethylene molecule occurs without the formation of significant amounts of other oxygenated compounds such as aldehydes. This novel reaction may be expressed as:

$$2CH_2{=}CH_2 + O_2 \longrightarrow 2CH_2{-}CH_2 \quad (O)$$

and it is accompanied by complete oxidation of some of the ethylene to give carbon dioxide and water according to the equation:

$$CH_2{=}CH_2 + 3O_2 \rightarrow 2CO_2 + 2H_2O$$

Ethylene from any desired source may be used, including that obtained from coke oven gas, hydrogenation of acetylene, or dehydration of ethyl alcohol. The oxygen can be the oxygen of the air, and can conveniently be mixed with the ethylene prior to the reaction.

The oxidation of ethylene takes place with a giving off of heat, and it is, of course, desirable to maintain the temperature of the zone of reaction within the range specified. This can be facilitated by suitable dilution of the reaction gases, such as that accomplished by the use of air as the source of oxygen, and some water or carbon dioxide in addition to that formed can be admitted to the mixture in the reaction zone if desired. Hydrogen may be similarly added.

The process can be conveniently conducted by passing a mixture containing air and ethylene through a tube, for example, of silica or of a material capable of withstanding high pressures, within which is placed such a surface catalyst as one composed of silver in finely divided form, and which may be disposed within the tube on a supporting material, or in other known ways. The temperature of the reaction zone is maintained between about 150° and about 400° C., for example, at about 200° C., and the issuing products from the tube contain ethylene oxide, carbon dioxide and water without objectionable aldehydes. The ethylene oxide can be recovered in any suitable way, preferably in liquid form by condensation or solution.

The surface catalyst is preferably silver as stated, but other metals such as bismuth and antimony may be used. Likewise oxides of these metals or mixtures containing these metals or their oxides may be used. Experiments have also indicated that the catalyst may be silver activated by the addition of small amounts of gold or copper or iron.

In any case, the ethylene and oxygen are thus reacted simultaneously at the temperatures set forth in the presence of a surface catalyst and of water, and the process is conducted to produce ethylene oxide as a direct product in substantial and economic amount. The ethylene oxide can be recovered by known methods, and it may be further reacted to form ethylene glycol or derivatives of ethylene glycol.

I claim:

1. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water at a temperature between 150° and 400° C. in the presence of a catalyst composed of silver activated by small quantities of a metal selected from a group consisting of gold, copper and iron.

2. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of the oxygen and water at a temperature between 150° and 400° C. in the presence of a catalyst composed of a mixture of silver, gold, copper and iron.

3. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water at a temperature between 150° C. and 400° C. in the presence of a catalyst composed of a mixture of silver and gold.

4. A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen and steam at a temperature between 150° and 400° C., and at a pressure below atmospheric pressure, in the presence of a catalyst composed of a metal selected from a group consisting of silver, gold, copper and iron, and in the presence of hydrogen in the proportion of 5 per cent of the total volume.

5. A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen and steam at a pressure below atmospheric pressure, in the presence of a catalyst composed of a metal selected from a group consisting of silver, gold, copper

and iron, and in the presence of hydrogen in the proportion of 5 per cent of the total volume, and of CO_2.

6. A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen at a temperature between 150° and 400° C. in the presence of a catalyst composed of silver activated with small quantities of gold.

7. A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen at a temperature between 150° and 400° C. in the presence of a catalyst composed of silver activated with small quantities of a mixture of gold and copper.

8. The process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene, which comprises forming a mixture containing ethylene and molecular oxygen and conducting said mixture through a confined reaction zone which is maintained at an elevated temperature; controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400° C.; subjecting said mixture in said zone at said elevated controlled temperature to intimate contact with an active surface catalyst of a material having the catalytic characteristics of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water and which does not favor the existence in said products of substantial amounts of aldehydes; correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide; and thereafter recovering ethylene oxide-containing product in liquid form.

9. The process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene, which comprises forming a mixture containing ethylene and molecular oxygen and a heat-absorbing diluent and conducting said mixture through a confined reaction zone which is maintained at an elevated temperature; controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400° C.; subjecting said mixture at said elevated controlled temperature to intimate contact with an active surface catalyst material composed essentially of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water and which does not favor the existence in said products of substantial amounts of aldehydes; correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide; and thereafter separating ethylene oxide from the products of oxidation.

THEODORE EMILE LEFORT.

PLAINTIFF'S EXHIBIT 7

DISTRICT COURT OF THE UNITED STATES

FOR THE DISTRICT OF MARYLAND.

CARBIDE AND CARBON CHEMICALS CORPORATION, *Plaintiff*, *vs.* U. S. INDUSTRIAL CHEMICALS, INC., *Defendant*.	Civil Action File No. 249

STATEMENT BY DEFENDANT.

In compliance with a request of counsel for plaintiff, defendant admits, for the purposes of this case as follows, and plaintiff accepts this admission in lieu of resort to other pre-trial discovery of acts of defendant alleged by plaintiff to constitute infringement of Reissue Letters Patent No. 20,370.

Subsequent to the grant of said patent, and after notice of plaintiff's asserted rights thereunder, and prior to the filing of the complaint herein, defendant engaged in the commercial production of ethylene glycol in its manufacturing plant at Baltimore, Maryland. In doing so, defendant employed a chamber containing a catalyst consisting of silver disposed upon a carrier of aluminum oxide bonded by a fusion process. A gaseous mixture containing ethylene and the constituents of atmospheric air was passed continuously through this chamber in intimate contact with said catalyst during an extended period of time.

The gaseous mixture entering the chamber contained usually about 1.5% by volume of water vapor, not exceeding 1.7% by volume at any time.

Throughout the operation the temperature within the said chamber was maintained between about 150° and about 400° C.

The ethylene and oxygen in the gaseous mixture reacted within the chamber, in the presence of the catalyst, to produce ethylene oxide in substantial amount; and the resultant gaseous mixture, containing the said oxide and no substantial amounts of aldehydes, passed out of the said chamber continuously during the operation. The said resultant gaseous mixture contained usually about 3.5% by volume of water vapor, not exceeding 3.7% by volume at any time.

Thereafter, the ethylene oxide was dissolved out of the said resultant gaseous mixture by passing said mixture into an acidified aqueous medium, and was reacted in the solution to form ethylene glycol.

L. A. WATSON
Counsel for Plaintiff,

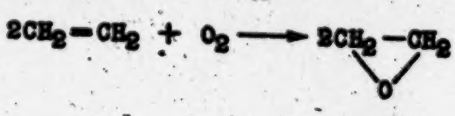
Dated: April 11, 1940

PENNIE, DAVIS, MARVIN & EDMONDS
Counsel for Defendant.

Defendant's Operation

"The ethylene and oxygen in the gaseous mixture reacted within the chamber, in the presence of the catalyst, to produce ethylene oxide . . ."

Thus there was direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene to make one molecule of ethylene oxide, according to the equation



"A gaseous mixture containing ethylene and the constituents of atmospheric air . . ."

The constituents of atmospheric air include molecular oxygen having the formula O_2.

"was passed continuously through this chamber . . ."

"Throughout the operation the temperature within said chamber was maintained between about 150° and about 400° C."

Said gaseous mixture was "in intimate contact with said catalyst" and "reacted within the chamber".

"a catalyst consisting of silver disposed upon a carrier of aluminum oxide bonded by a fusion process".

"The ethylene and oxygen in the gaseous mixture reacted within the chamber, in the presence of the catalyst, to produce ethylene oxide in substantial amount; and the resultant gaseous mixture, containing the said oxide and no substantial amounts of aldehydes, passed out of said chamber continuously during the operation."

"The said resultant gaseous mixture contained usually about 3.5% by volume of water vapor,"

"The gaseous mixture entering the chamber contained usually about 1.5% by volume of water vapor,"

Said gaseous mixture "was passed continuously through this chamber in intimate contact with said catalyst during an extended period of time".

"Throughout the operation the temperature within the said chamber was maintained between about 150° and about 400° C."

"The ethylene and oxygen in the gaseous mixture reacted within the chamber, in the presence of the catalyst, to produce ethylene oxide in substantial amount;"

"Thereafter, the ethylene oxide was dissolved out of said resultant gaseous mixture by passing said mixture into an acidified aqueous medium, and was reacted in the solution to form ethylene glycol".

This medium was in liquid form and contained the ethylene oxide.

Plaintiff's Exhibit 8

CLAIM 8 of Reissue 20,370

The process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene,

which comprises forming a mixture containing ethylene and molecular oxygen

and conducting said mixture through a confined reaction zone

which is maintained at an elevated temperature; controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400° C.;

subjecting said mixture in said zone at said elevated controlled temperature to intimate contact with an active surface catalyst

of a material having the catalytic characteristics of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water and which does not favor the existence in said products of substantial amounts of aldehydes;

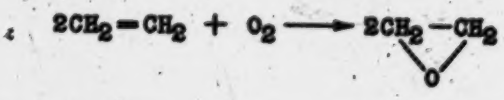
correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide;

and thereafter recovering ethylene oxide-containing product in liquid form.

Defendant's Operation

"The ethylene and oxygen in the gaseous mixture reacted within the chamber, in the presence of the catalyst, to produce ethylene oxide . . ."

Thus there was direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene to make one molecule of ethylene oxide, according to the equation



"A gaseous mixture containing ethylene and the constituents of atmospheric air . . ."

"The gaseous mixture entering the chamber contained usually about 1.5% by volume of water vapor,"

The constituents of atmospheric air include molecular oxygen having the formula O_2 and substantial amounts of nitrogen, a diluent.

"was passed continuously through this chamber . . ."

"Throughout the operation the temperature within said chamber was maintained between about 150° and about 400° C."

Said gaseous mixture was "in intimate contact with said catalyst" and "reacted within the chamber".

"a catalyst consisting of silver disposed upon a carrier of aluminum oxide bonded by a fusion process".

"The ethylene and oxygen in the gaseous mixture reacted within the chamber, in the presence of the catalyst, to produce ethylene oxide in substantial amount; and the resultant gaseous mixture, containing the said oxide and no substantial amounts of aldehydes, passed out of said chamber continuously during the operation."

"The said resulfant gaseous mixture contained usually about 3.5% by volume of water vapor,"

"The gaseous mixture entering the chamber contained usually about 1.5% by volume of water vapor,"

Said gaseous mixture "was passed continuously through this chamber in intimate contact with said catalyst during an extended period of time".

"Throughout the operation the temperature within the said chamber was maintained between about 150° and about 400° C."

"The ethylene and oxygen in the gaseous mixture reacted within the chamber, in the presence of the catalyst, to produce ethylene oxide in substantial amount;"

"Thereafter, the ethylene oxide was dissolved out of said resultant gaseous mixture by passing said mixture into an acidified aqueous medium, and was reacted in the solution to form ethylene glycol".

Plaintiff's Exhibit 9

CLAIM 9 of Reissue 20,370

The process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene,

which comprises forming a mixture containing ethylene and molecular oxygen and a heat-absorbing diluent

and conducting said mixture through a confined reaction zone

which is maintained at an elevated temperature; controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400° C.;

subjecting said mixture in said zone at said elevated controlled temperature to intimate contact with an active surface catalyst

material composed essentially of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water and which does not favor the existence in said products of substantial amounts of aldehydes;

correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide;

and thereafter separating ethylene oxide from the products of oxidation.

Patented Apr. 23, 1935

1,998,878

UNITED STATES PATENT OFFICE

1,998,878

PROCESS FOR THE PRODUCTION OF ETHYLENE OXIDE

Théodore Emile Lefort, Paris, France, assignor to Société Anonyme; Societe Francaise de Catalyse Generalisee, Paris, France

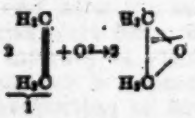
No Drawing. Application March 22, 1932, Serial No. 600,578. In France March 27, 1931

7 Claims. (Cl. 260—156.5)

REISSUED

This invention has for object a process for the production of ethylene oxide which mainly consists in subjecting ethylene to the simultaneous action of the oxygen of air and of water, in presence of a catalyzer and, if need be, of hydrogen.

Hydrogen, in case it is employed, in contact with the catalyzer, probably plays the part of an agent for accelerating the reaction which can be written as follows:



This reaction takes place at atmospheric pressure but, for commercially carrying the said process into practice, pressure can be employed, for instance a pressure of 50 kilograms per square centimeter, at a temperature between 150 and 400° C.

The following catalyzers may be used:

(1) A mixture of antimony, bismuth, silver, lead, arsenic, nickel and tin, in variable proportions.

(2) Silver alone.

(3) Silver activated by introducing therein small proportions of gold, copper or iron.

(4) The metals: gold, copper, iron, separately or in admixture and in any proportions.

(5) Silver alone, bismuth alone and antimony alone.

(6) The following mixtures:

Silver, gold, copper, iron	in any proportions
Silver, gold, copper	
Silver, gold	
Silver, copper, iron	
Silver, copper	
Silver, iron	
Copper, iron	

(7) The mixtures of antimony, lead, nickel and silver.

(8) The mixtures of bismuth, nickel, silver and lead.

(9) The mixtures of antimony, bismuth, nickel.

(10) The mixtures of antimony, bismuth, silver.

(11) The mixtures of bismuth and nickel.

(12) The mixtures of bismuth and silver.

And, generally speaking, all the mixtures which can be obtained with the following metals: antimony, lead, bismuth, silver, nickel, tin, arsenic, gold, copper, iron, taken separately, taken two by two, three by three, four by four, etc. . . . in variable proportions, the mixtures, in the same conditions, of these metals with their oxides, the mixtures of the oxides alone of these metals, in the same conditions.

The ethylene can be obtained from any source of supply: gas from coke ovens, hydrogenation of acetylene, dehydration of ethyl alcohol, and the like.

Water can be admitted in the reaction vessel, either in the liquid state, or as steam.

The oxygen can be the oxygen of the air, this latter gas being introduced in the reaction.

The efficiency of the reaction is increased by diminishing the CO_2 which is formed, by introduction, in this reaction, of a suitable quantity of water: A suitable volume of CO_2 can also be previously introduced in the reacting gases.

For increasing the yield in ethylene oxide, the process can also be carried out under a pressure lower than atmospheric pressure.

Experiments have shown that silver could be activated by introducing, in this body, small quantities either of gold, or of copper, or of iron.

Thus, a mixture containing silver, gold, copper and iron, gives very satisfactory results and allows a specific reaction; the only foreign product obtained in the reaction is CO_2 which can be easily removed. Aldehydes formed exist only as traces.

Moreover, the applicant has found that the reaction giving CO_2 is, contrarily to previous belief, a reaction of oxidation independent from that giving ethylene oxide and from that giving aldehydes. From experiments effected by the applicant, it results that, if water is introduced in suitable quantity, the reaction is not only facilitated, as above stated, but, in addition, the reaction giving CO_2 probably by direct oxidation of ethylene according to the equation:

$$C_2H_4+3O_2=2H_2O+2CO_2$$

is checked, owing, as is probable, to the partial pressure of water. From these experiments, it can be concluded that if a certain quantity of CO_2 is previously introduced, in addition, in the reacting gases, the partial pressure of CO_2 will produce its effect, and the reaction of oxidation of ethylene into CO_2 will be completely checked resulting in a considerable increase in the efficiency of the operation. Moreover, it is to be noted that, when the synthetic operation is effected under partial vacuum, the yield in ethylene oxide is increased.

Several modi operandi for carrying the process

into practice, will be indicated hereinafter, by way of example only.

Modus operandi I

In a plant constructed for resisting to high pressures, ethylene on the one hand, and air on the other hand, are separately compressed by means of suitable compressors.

The mixture of air and ethylene can also be compressed by taking the necessary precautions in order to avoid the risk of explosion.

These two gases will be led, together with hydrogen or not, into a reaction tube resisting to the pressure of several hundreds of kilograms, containing one of the catalysers indicated above, and heated, for instance, to a temperature of 200° C. This tube communicated moreover with a circulating pump allowing to introduce water under pressure.

The reaction will take place in this tube between ethylene, the oxygen of air, and water. By controlling the outputs, a continuous operation will be obtained. Ethylene oxide thus formed will be separated from the other gases, for instance, by condensation.

Modus operandi II

Silver in powder form, or in the form of a wire gauze, and placed or not on a support or carrier, is introduced in a tube, made for instance of silica, and, after the said tube has been heated to a temperature comprised between 150 and 400° C., a mixture of ethylene, air, water vapour, and hydrogen is caused to pass therethrough. The proportion of hydrogen is about 5% of the total mixture. The ethylene oxide produced can be separated from the gases issuing from the tube.

Modus operandi III

A silver colloid is introduced in a high pressure tube which has been filled with water. Pure ethylene under pressure is then added in order that it can dissolve in the water.

The tube is subsequently heated between 150 and 400° C., then air and hydrogen are slowly introduced. The introduction of air is stopped when the totality of ethylene is converted into ethylene oxide.

From ethylene oxide produced according to the processes which have been indicated above, it is possible to obtain, by simple saponification, ethylene-glycol, or by other known methods, any derivative of ethylene-glycol.

The reaction, for obtaining ethylene-glycol, can be written as follows:

$$C_2H_4O + H_2O \rightarrow \begin{matrix} H_2C-OH \\ H_2C-OH \end{matrix}$$

The experiments effected by the applicant have shown that, in presence of the catalysers indicated, water, in the form of steam or not, considerably promotes the reaction ensuring the production of ethylene oxide.

What I claim as my invention and desire to secure by Letters Patent is:

1. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst, constituted by silver activated by introducing in the body small quantities of a metal selected from a group consisting of gold, copper, and iron, at a temperature between 150 and 400° C.

2. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of the oxygen and water, in presence of a catalyst, constituted by a mixture of silver, gold, copper and iron, at a temperature between 150 and 400 degrees C.

3. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst constituted by a mixture of silver and gold, at a temperature between 150 and 400° C.

4. A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen and steam, in presence of a catalyst constituted by a metal selected from a group consisting of silver, gold, copper and iron, and hydrogen in the proportion of 5 per cent of the total volume, at a temperature between 150 and 400 degrees C., and at a pressure below atmospheric pressure.

5. A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen and steam, in presence of a catalyst constituted by a metal selected from a group consisting of silver, gold, copper and iron, and hydrogen in the proportion of 5 per cent of the total volume, and CO_2, and at a pressure below atmospheric pressure.

6. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen in presence of a catalyst constituted by silver activated with small quantities of gold, at a temperature between 150 and 400 degrees C.

7. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen in presence of a catalyst constituted by silver activated with small quantities of a mixture of gold and copper at a temperature between 150 and 400 degrees C.

THEODORE EMILE LEFORT.

Defendant's Exhibit 3

285

ETATS-UNIS D'AMERIQUE

600578

REGD. MAR 31 1932 DIVISION 8

PETITION.

TO THE COMMISSIONNER OF PATENTS :

Your petitioner Théodore Emile LEFORT *residing at* PARIS (France)

a *citizen* / ~~subject~~ } *of* the Republic of France

and whose Post Office Address is 8 Avenue Percier, PARIS (France)

prays that Letters Patent may be granted to him *for the improvement* in a process for the production of ethylene oxide, particularly for the preparation of ethylene-glycol and of its derivatives *set forth in the annexed Specification.*

INSERT

And he hereby appoint s Marks & Clerk a firm Composed of G. C. Marks, P. Clerk & M. A. Adam, national Press Bldg. Washington, D.C. his

Attorneys with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign the drawings, to receive the Letters Patent, and to transact all business in the United States Patent Office connected therewith.

Signed at Paris *this* 11th March 1932

Théodore Emile Lefort (1) *Signature*

SPECIFICATION.

TO ALL WHOM IT MAY CONCERN :

BE IT KNOWN that I, Théodore Emile LEFORT

a *citizen* / ~~subject~~ } *of* the Republic of France

residing at PARIS (France)

have invented certain new and useful improvements in a process for the production of ethylene oxide, ~~particularly for the preparation of ethylene-glycol and of its derivatives~~

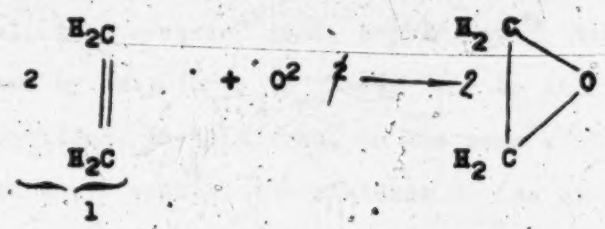
of which the following is a Specification :

600578

1

This invention has for object a process for the production of ethylene oxide which mainly consists in subjecting ethylene to the simultaneous action of the oxygen of air and of water, in presence of a catalyser and, if need be, of hydrogen.

Hydrogen, in case it is employed, in contact with the catalyser, probably plays the part of an agent for accelerating the reaction which can be written as follows :



This reaction takes place at atmospheric pressure but, for commercially carrying the said process into practice, pressure can be employed, for instance a pressure of 50 kilograms per square centimeter, at a temperature between 150 and 400° C.

The following catalysers may be used :

(1°) A mixture of antimony, bismuth, silver, lead, arsenic, nickel and tin, in variable proportions.

(2°) Silver alone.

(3°) Silver activated by introducing therein small proportions of gold, copper or iron.

(4°) The metals : gold, copper, iron, separately or in admixture and in any proportions.

(5°) Silver alone, bismuth alone and antimony alone.

(6°) The following mixtures :

- Silver, gold, copper, iron,
- Silver, gold, copper,
- Silver, gold,
- Silver, copper, iron,
- Silver, copper,
- Silver, iron,
- Copper, iron,

} in any proportions

(7°) The mixtures of antimony, lead, nickel and silver,

(8°) The mixtures of bismuth, nickel, silver and lead.

(9°) The mixtures of antimony, bismuth, nickel.

(10°) The mixtures of antimony, bismuth, silver.

(11°) The mixtures of bismuth and nickel.

(12°) The mixtures of bismuth and silver.

And, generally speaking, all the mixtures which can be obtained with the following metals : antimony, lead, bismuth, silver, nickel, tin, arsenic, gold, copper, iron, taken separately, taken two by two, three by three, four by four, etc... in variable proportions, the mixtures, in the same conditions, of these metals with their oxides, the mixtures of the oxides alone of these metals, in the same conditions.

The ethylene can be obtained from any source of supply : gas from coke ovens, hydrogenation of acteylene, dehydration of ethyl alcohol, and the like.

Water can be admitted in the reaction vessel, either in the liquid state, or as steam.

The oxygen can be the oxygen of the air, this latter gas being introduced in the reaction.

The efficiency of the reaction is increased by diminishing the CO_2 which is formed, by introduction, in this reaction, of a suitable quantity of water. A suitable volume of CO_2 can also be previously introduced in the reacting gases.

For increasing the yield in ethylene oxide, the process can also be carried out under a pressure lower than atmospheric pressure.

Experiments have shown that silver could be activated by introducing, in this body, small quantities either of gold, or of copper, or of iron.

Thus, a mixture containing silver, gold, copper and iron, gives very satisfactory results and allows a specific reaction; the only foreign product obtained in the reaction is CO_2 which can be easily removed. Aldehydes formed exist only as traces.

Defendant's Exhibit 3 (Continued)

Moreover, the Applicant has found that the reaction giving CO_2 is, contrarily to previous belief, a reaction of oxidation independent from that giving ethylene oxide and from that giving aldehydes. From experiments effected by the Applicant, it results that, if water is introduced in suitable quantity, the reaction is not only facilitated, as above stated, but, in addition, the reaction giving CO_2, probably by direct oxidation of ethylene according to the equation :

$$C_2H_4 + 3\ O_2 = 2\ H_2O + 2\ CO_2$$

is checked, owing, as is probable, to the partial pressure of water. From these experiments, it can be concluded that if a certain quantity of CO_2 is previously introduced, in addition, in the reacting gases, the partial pressure of CO_2 will produce its effect, and the reaction of oxidation of ethylene into CO_2 will be completely checked resulting in a considerable increase in the efficiency of the operation. Moreover, it is to be noted that, when the synthetic operation is effected under partial vacuum, the yield in ethylene oxide is increased.

Several modi operandi for carrying the process into practice, will be indicated hereinafter, by way of example only.

MODUS OPERANDI

In a plant constructed for resisting to high pressures, ethylene on the one hand, and air on the other hand, are separately compressed by means of suitable compressors.

The mixture of air and ethylene can also be compressed by taking the necessary precautions in order to avoid the risk of explosion.

These two gases will be led, together with hydrogen or not, into a reaction tube resisting to the pressure of several hundreds of kilograms, containing one of the catalysers indicated above, and heated, for instance, to a temperature of 200 ° C. This tube communicated moreover with a circulating pump, allowing to introduce water under pressure.

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The reaction will take place in this tube between ethylene, the oxygen of air, and water. By controlling the outputs, a continuous operation will be obtained. Ethylene oxide thus formed will be separated from the other gases, for instance, by condensation.

MODUS OPERANDI II

Silver in powder form, or in the form of a wire gauze, and placed or not on a support or carrier, is introduced in a tube, made for instance of silica, and, after the said tube has been heated to a temperature comprised between 150 and 400° C, a mixture of ethylene, air, water vapour, and hydrogen is caused to pass therethrough. The proportion of hydrogen is about 5 % of the total mixture. The ethylene oxide produced can be separated from the gases issuing from the tube.

MODUS OPERANDI III

A silver colloid is introduced in a high pressure tube which has been filled with water. Pure ethylene under pressure is then added in order that it can dissolve in the water.

The tube is subsequently heated between 150 and 400° C, then air and hydrogen are slowly introduced. The introduction of air is stopped when the totality of ethylene is converted into ethylene oxide.

From ethylene oxide produced according to the processes which have been indicated above, it is possible to obtain, by simple saponification, ethylene-glycol, or by other known methods, a derivative of ethylene-glycol.

The reaction, for obtaining ethylene-glycol, can be written as follows :

$$C_2H_4O + H_2O \longrightarrow \begin{matrix} H_2C - OH \\ | \\ H_2C - OH \end{matrix}$$

The experiments effected by the Applicant have shown that, in presence of the catalysers indicated, water, in the form of steam or not, considerably promotes the reaction ensuring the production of ethylene oxide.

What I claim as my invention and desire to secure by Letters Patent is :

1. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneousaction of oxygen and water, in presence of a catalyst, and, if need be, of hydrogen.

2. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst, constituted by a mixture of antimony, bismuth, silver, lead, arsenic, nickel and tin, at a temperature between 150 and 400° C.

3. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst, constituted by silver, in a powder state or in the shape of a wire-gauze, at a temperature between 150 and 400° C.

4. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst, constituted by silver activated by introducing in the body, small quantities either of gold, or of copper, or of iron, at a temperature between 150 and 400° C.

5. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst constituted by gold, silver, iron alone or mixed, at a temperature between 150 and 400° C.

6. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of the oxygen and water, in presence of a catalyst, constituted by a mixture of silver, gold, copper and iron, at a temperature between 150 and 400 degrees C.

7. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst constituted by a mixture of silver, gold and copper, at a temperature between 150 and 400° C.

8. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst constituted by a mixture of silver and gold, at a temperature between 150 and 400 ° C.

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9. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst, constituted by a mixture of silver, copper and iron, at a temperature between 150 and 400° C.

10. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst constituted by a mixture of silver and copper, at a temperature between 150 and 400 ° C.

11. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst constituted by a mixture of silver and iron, at a temperature between 150 and 400 ° C.

12. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water, in presence of a catalyst constituted by a mixture of copper and iron, at a temperature between 150 and 400° C.

~~13.~~ A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen and steam, in presence of a catalyst and hydrogen in the proportion of 5 per cent of the total volume, at a temperature between 150 and 400 degrees C. and at pressure below atmospheric pressure.

~~14.~~ A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen and steam, in presence of a catalyst and hydrogen in the proportion of 5 per cent of the total volume, and CO_2, and at pressure below atmospheric pressure.

600578

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ETATS-UNIS D'AMERIQUE

B 292

In testimony whereof I *have signed this specification*

Théodore Emile Lefort (2) *Signature*

OATH.

CONSULATE GENERAL OF THE U.S.A. }
} ss.
}

Théodore Emile LEFORT

the above-named petitioner , being duly sworn or affirmed depose s and say s that he is a *citizen subject* of the Republic of France *and resident of* 8 Avenue Percier, PARIS (France) *that* he *verily believes* himself *to be the original, first, and* sole *inventor of the improvements in* a process for the production of ethylene oxide, particularly for the preparation of ethylene-glycol and of its derivatives *described and claimed in the annexed specification; that* he does *not know and do es not believe that the same was ever known or used before* his *invention or discovery thereof, or patented or described in any printed publication in any country before* his *invention or discovery thereof, or more than two years prior to this application, or in public use or on sale in the United States for more than two years prior to this application; that said invention has not been patented in any country foreign to the United States on an application filed by* him *or* his *legal representatives or assigns more than twelve months prior to this application; and that no application for patent on said improvement has been filed by* him *or* his *representatives or assigns in any country foreign to the United States except as follows*

- in France on the 27th March 1931,
- " " " " 4th July 1931 (Patent of addition)
- " " " " 11th September 1931 (Patent of Addition)
- " " " " 28th September 1931 (Patent of Addition)
- " " " " 19th December 1931 (Patent of Addition)
- " " " " 3rd October 1931,
- " " " " 19th December 1931 (Patent of Addition)

Théodore Emile Lefort (3) *Signature*

SWORN to and subscribed before me this 11th *day of* March 1932

2398

[redacted] (4)
Vice-Consul of the United States
U. S. Consul

AMERICAN CONSULATE
PARIS

600578

8

8.

Div. 6 Room 4898C ca/ah Paper No. 2

Address only "The Commissioner of Patents, Washington, D. C." and not any official by name

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
WASHINGTON

All communications respecting this application should give the serial number, date of filing, and name of the applicant

Sept. 16, 1932

Please find below a communication from the EXAMINER in charge of this application.

Thomas E. Robertson
Commissioner of Patents.

Applicant: T. E. Lefort

Marks & Clerk
National Press Bldg.
Washington, D. C.

Ser. No. 600,578
Filed Mar. 22, 1932
For Process for the Production of Ethylene Oxide, Particularly for the Preparation of Etc.

MAILED
SEP 16 1932

Examined.

The title should be amended to indicate that applicant's invention resides in the production of ethylene oxide. The hydrolysis of the oxide to ethylene glycol forms no part of applicant's invention, since it is obvious that the oxide derived from any source could be hydrolized.

Division and election of species between the following groups of claims is required in accordance with the provisions of Rule 41:

I. Claim 2 in which a mixture composed of antimony, bismuth, silver, lead, arsenic, nickel and tin is used as a catalyst.

II. Claim 3 in which silver is used.

III. Claim 6 in which a mixture composed of silver, gold, copper and iron is used.

IV. Claim 7 in which a mixture composed of silver, gold and copper is used.

V. Claim 8 in which a mixture composed of silver and gold is used.

VI. Claim 9 in which a mixture composed of silver, copper and iron is used.

VII. Claim 10 in which a mixture composed of silver and copper is used.

VIII. Claim 11 in which a mixture composed of silver and iron is used.

600578

9

Serial No. 600,578 - #2.

IX. Claim 12 in which a mixture composed of copper and iron is used.

Claims 1, 13 and 14 are generic to all of the above groups. Claim 4 is generic to groups V, VII and VIII. Claim 5 is generic to group II. However, claims 1, 13 and 14 are rejected as being indefinite and too broad in "catalyst" and claims 4 and 5 are rejected as being alternative in "either of gold, or of copper or of iron" and "alone or mixed". Alternative expressions are not allowed in patent claims. The term "catalyst" does not identify the materials used and is nothing more than a statement of a desired result or affect. There is no invention in the broad idea of using a catalyst in any process.

In the absence of an allowable generic claim only one specie may be claimed. As to a proper response to the requirement for division, if the patentability of the generic claims is urged, applicant's attention is directed to Ex parte Doane, 410 O. G. 275.

W.H.C

A M Lewers
Examiner.

600578 #10

AMENDMENT

IN THE UNITED STATES PATENT OFFICE

T. E. Lefort
Serial No. 600578
Filed March 22, 1932
For: Process for the Production of Ethylene Oxide, Particularly for the Preparation of Ethylene-glycol etc.
Division 6....Room 4808C

Hon. Commissioner of Patents,

Washington, D.C.

Sir:

In response to the official letter of Sept. 16, 1932, please amend the above case as follows:

Change the title of the invention to read: --Process for the Production of Ethylene Oxide.--

Cancel claims 2, 3, 9, 10, 11 and 12.

Claim 1, change the last line to read --and water, in presence of a catalyst, constituted by metal selected from a group consisting of silver, gold, copper and iron, and in presence or hydrogen.--

Claim 4, line 4, change "either" to--of metal selected from a group consisting--. Same line, cancel "or of". Last line, change ", or of" to --and--.

Claim 5, line 3, before "gold" insert --metal selected from a group consisting of--. Same line, change the comma after "silver" to --and--. Last line, cancel "alone or mixed".

Claim 13, line 3, after "catalyst" insert --constituted by metal selected from a group consisting of silver, gold, copper and iron,--.

Claim 14, line 3, after "catalyst" insert --constituted by metal selected from a group consisting or silver, gold, copper and iron,--.

Claim 8, line 3, after "silver" insert --and--?

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- R E M A R K S -

The requested division has now been made by cancelling claims 2, 3, 9, 10, 11 and 12. Of the remaining claims, 1, 4, 6, 13 and 14 are generic and three claims, 5, 7 and 8 are specific. The alternatives have been removed and other suitable corrections made in the standing claims. The catalyst has been specified in each of the claims now remaining.

A favorable action is solicited.

Respectfully submitted,

By

Marks & Clerk

Attorneys.

March 15, 1933

GHJ-ED-17168

600578

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12.

SUPPLEMENTAL AMENDMENT

IN THE UNITED STATES PATENT OFFICE

T.E.LEFORT
SER.NO.600578
FILED MARCH 22, 1932
FOR: PROCESS FOR THE PRODUCTION OF ETHYLENE OXIDE, PARTICULARLY FOR THE PREPARATION OF ETHYLENE-GLYCOL ETC.
DIVISION 6 ROOM 4898C

Hon. Commissioner of Patents,

Washington, D.C.

Sir:

In further response to the official action of September 15, 1932, please add the following claims:

--15. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen in presence of a catalyst constituted by silver activated with small quantities of another metal, at a temperature between 150 and 400 degrees C.

16. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen in presence of a catalyst constituted by silver activated with small quantities of gold, at a temperature between 150 and 400 degrees C.

17. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen in presence of a catalyst constituted by silver activated with small quantities of copper, at a temperature between 150 and 400 degrees C.

SC 05718 - 4/13

18. A process for the production or ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen in presence of a catalyst constituted by silver activated with small quantities of a mixture of gold and copper at a temperature between 150 and 400 degrees C.

19. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen in presence of a catalyst constituted by silver activated with small quantities of another metal selected from group I of the periodic system at a temperature between 150 and 400 degrees C. and at a pressure below atmospheric pressure, the proportion of hydrogen being of 5 per cent or the total volume of oxygen and steam. --

- R E M A R K S -

It is respectfully requested that the added claims be considered with the previous amendment.

Respectfully submitted,

By Marks & Clerk
Attorneys.

March.21, 1933

GHJ-ED-17168

600578

Div. 6 Room 4898 C

Paper No. 5

Address only "The Commissioner of Patents, Washington, D. C." and not any official by name

All communications respecting this application should give the serial number, date of filing, and name of the applicant

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
WASHINGTON

GA:RW

June 24, 1933.

Please find below a communication from the EXAMINER in charge of this application.

Thomas E. Robertson
Commissioner of Patents.

Applicant: T. E. Lefort

Marks & Clerk,
National Press Bldg.,
Washington, D. C.

Ser. No. 600,578
Filed Mar. 22, 1932
For Process For The Production Of Ethylene Oxide.

MAILED
JUN 24 1933

In response to amendments filed Mar. 16 and 23, 1933.

Additional references:

Skarbloom (British) 369,141 acc. Mar. 17, 1932 260-156.5
Article by Lenher "Jour. Am. Chem. Soc." Vol 53, pages 2420 to 2421 (1931)
Article by Willstatter, "annalen des Chemie" Vol. 422, pages 36 to 46 (1920).

Claims 1 and 5 are rejected upon Willstatter in view of Lenher. Both references disclose the oxidation of ethylene under the same conditions as are used by the applicant. Willstatter discloses the use of metal catalysts such as copper, silver, etc. Lenher discloses that ethylene oxide is one of the products formed. Willstatter discloses the use of inert diluents and the use of steam as a diluent is not therefore considered to involve invention. Skarbloom discloses a similar processes in which the steam is used as a diluent. Willstatter also discloses that steam is present during the reaction.

Claims 4, 5, 13 and 14 are unidiomatic in "by metal". This should be ---by metals---. "Metal" alone is not good English.

Claims 15 and 19 are rejected as being too broad in "another metal". Applicant has not disclosed that all other metals may be used as activators. The claims must be commensurate with the disclosure.

Division is required between the following groups of claims:

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SHEET NO. 2 Serial No. 600,578

(I) claim 6, catalyst compound of Ag, An, Cu and Fe,

(II) claims 7 and 17, catalyst compound of Ag, An and Cu.,

(III) Claims 8 and 15, catalyst compound of Ag and An; and

(IV) Claim 16, catalyst compound of Ag and Cu.

Not more than three species can be had with an allowable generic claim and four are claimed in this case.

W.H.C.

A M Sievers

EXAMINER.

C

AMENDMENT

IN THE UNITED STATES PATENT OFFICE

T.E. Lefort
Serial No. 600578
Filed March 22,1932
Process for the Production of
Ethylene Oxide.
Division 6....Room 4898C.

Hon. Commissioner of Patents,

Washington, D.C.

Sir:

In response to the official letter of June 24, 1933, please amend the above case as follows:

Cancel claims 1, 5, 7 and 17.

Claims 15 and 19, line 5, after "metal" insert --selected from group I of the periodic system--.

- R E M A R K S -

The requested division has now been made by cancellation of claims 7 and 17 so that only three species are now claimed beside the generic claims 13 and 14.

Claims 1 and 5 which appear to be met by the cited references have been cancelled.

With regard to the requested correction of claims 4, 5, 13 and 14 to change "metal" to -metals-, this change has not been made because it is considered that the claims read better in their present state than they would after such change. In connection herewith it is pointed out that the Markush decision has been applied in said claims in order to avoid the alternative expression "either of gold or of copper or of iron" which occurs in said claims. It will further be noted that the word

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"metal" in this case is collective in order to imply that not only one but two or more of the enumerated metals may be used and this is considered perfectly good English. Since no references have been cited against the now standing claims namely, 4, 6, 8, 13, 14, 15, 16, 18 and 19, a favorable action passing the case to issue is solicited.

Respectfully submitted,

By Marbaugh Clark

Attorneys

December 22,1933

GHJ-ED- 17168

600578 / 18

18.

Defendant's Exhibit 3 (Continued)

303

JAN 5 1934
DIVISION 6

JAN-3-34

Letter

SUPPLEMENTAL AMENDMENT

IN THE UNITED STATES PATENT OFFICE.

T.E.Lefort
Serial No. 600578
Filed March 22, 1932
For: Process for the Production of Ethylene Oxide
Division 6....Room 4898C

Hon. Commissioner of Patents,

Washington, D.C.

Sir:

Please consider the following arguments in connection with the amendment filed Dec. 22, 1933:

The invention defined in the claims now maintained consists in a process of direct oxidation of the ethylene in the presence of oxygen, water (or steam) and hydrogen in the presence of a well-defined catalyst; the silver is activated with small quantities of gold, copper or iron.

The British Patent 369141 cannot be considered a prior publication. It was published on 17th March 1932 while applicant filed the present U.S. application under the Convention with priorities of March 27, 1931.

Lenher shows the secondary reactions which applicant proposes to eliminate in order to increase the yield of ethylene oxide. The catalysts cited by Willstatter do not include silver activated with small quantities of other metals.

A favorable action is solicited.

Respectfully submitted,

By Marrs & Clark
Attorneys.

December 20,1933
GEJ-ED-17163

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+3/9

Defendant's Exhibit 3 (Continued)

304

Address only "The Commissioner of Patents, Washington, D. C." and not any official by name

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
WASHINGTON

Papers No. ___

All communications regarding this application should give the serial number, date of filing, and name of the applicant

Please find below a communication from the EXAMINER in charge of this application.

Commissioner of Patents.

Mar. 28, 1934

Applicant: T. E. Lefort

Ser. No. 600,578
Filed Mar. 22, 1932
For Process for the Production of Ethylene Oxide

Marks & Clerk
National Press Bldg.
Washington, D. C.

MAR 28 1934

Responsive to amendment filed Dec. 22, 1933 and latter filed Jan. 3, 1934, respectively.

Claims 15 and 19 are rejected upon the ground that they have no basis in the disclosure. There was no disclosure that all metals of group I could be used. The metals of this group differ greatly in properties and the disclosure of a few could in no way indicate that all would be useful. Catalysis cannot be predicted.

The requirement that "metal" or "by metal", occurring in claims 4, 13 and 14, be changed to a metal or similar idiomatic English is repeated.

Claims 6, 8, 16 and 18 are allowable so far as now known.

W.H.C.

O. F. Setearns
Acty. Examiner.

600578

20.

IN THE UNITED STATES PATENT OFFICE

In re application

Applicant: T.E.Lefort

Title: Process for the Production of Ethylene Oxide

Ser. No.: 600578

Filed: March 22, 1932

Div. 6 Room 4898C

SUBSTITUTE POWER-OF-ATTORNEY

Hon. Commissioner of Patents,

Washington, D. C.

Sir:

In the matter of the above patent application please substitute Eustace S. Glascock, Wm. Webster Downing and Charles P. Seebold, constituting the firm of Glascock, Downing & Seebold (whose register number is 13992), National Press Building, Washington, D. C., as attorneys in our stead.

Marshall Celeste

September 17, 1934

600578

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AMENDMENT

IN THE UNITED STATES PATENT OFFICE

T. E. Lefort
Serial No. 600578
Filed March 22, 1932
For: Process for the Production of Ethylene Oxide
Division 6...Room 4898C

Hon. Commissioner of Patents,

Washington, D.C.

Sir:

In response to the official letter of March 28, 1934, please amend the above case as follows:

Cancel claims 15 and 19.

Claim 4, rewrite line 4 to read by introducing in the body small quantities of a metal selected from a group consisting of gold,

Claim 13, change line 3 to read gen and steam, in presence of a catalyst constituted by a metal selected from a group consisting of silver, gold, copper and iron, and hydrogen in the pro-

Claim 14, change line 3 to read and steam, in presence of a catalyst constituted by a metal selected from a group consisting of silver, gold, copper and iron, and hydrogen in the proportion

- R E M A R K S -

The rejected claims 15 and 19 having been cancelled and 4, 13 and 14 amended as directed by the Examiner, the case is now considered to be in condition for allowance since claims 6, 8, 16 and 18 have already been declared allowable. The standing and allowed claims are

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accordingly the following: 4, 6, 8, 13, 14, 16 and 18.

An early final action passing the case to issue is respectfully requested.

Respectfully submitted,

By Glassock, Downing & Seebold
Attorneys.

September 17, 1934
GHJ-ED-17168

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ADDRESS ONLY
THE COMMISSIONER OF PATENTS
WASHINGTON, D. C.
Div. 6-AH

Serial No. 600,578

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
WASHINGTON January three, 1935

Theodore Emile Lefort, Assor.,

Your APPLICATION for a patent for an IMPROVEMENT in Process for the Production of Ethylene Oxide

filed Mar. 22, 1932 has been examined and ALLOWED with 7 claims.

The final fee, THIRTY DOLLARS, WITH $1 ADDITIONAL FOR EACH CLAIM ALLOWED IN EXCESS OF 20, must be paid not later than SIX MONTHS from the date of this present notice of allowance. If the final fee be not paid within that period, the patent will be withheld, but the application may be renewed within one year after the date of the original notice with a renewal fee of $30 and $1 additional for each claim in excess of 20.

The office delivers patents upon the day of their date, on which date their term begins to run. The preparation of the patent for final signing and sealing will require about four weeks, and such work will not be begun until after payment of the necessary final fee.

When the final fee is paid, there should also be sent, DISTINCTLY AND PLAINLY WRITTEN, the name of the INVENTOR, TITLE OF THE INVENTION, AND SERIAL NUMBER AS ABOVE GIVEN, DATE OF ALLOWANCE (which is the date of this circular), DATE OF FILING, and, if assigned, the NAMES OF THE ASSIGNEES.

If it is desired to have the patent issue to an ASSIGNEE OR ASSIGNEES, an assignment containing a REQUEST to that effect, together with the FEE for recording the same, must be filed in this office on or before the date of payment of the final fee.

After issue of the patent, uncertified copies of the drawings and specifications may be purchased at the price of TEN CENTS EACH. The money should accompany the order. Postage stamps will not be received.

The final fee will NOT be received from other than the applicant, his assignee or attorney, or a party in interest as shown by the records of the Patent Office.

NOTICE.—WHEN THE NUMBER OF CLAIMS ALLOWED IS IN EXCESS OF 20, NO SUM LESS THAN $30 PLUS $1 ADDITIONAL FOR EACH CLAIM IN EXCESS OF TWENTY CAN BE ACCEPTED AS THE FINAL FEE.

Respectfully,

Commissioner of Patents.

Glascock, Downing & Seebold
National Press Bldg.
Washington, D. C.

600578

24.

IN REMITTING THE FINAL FEE GIVE THE SERIAL NUMBER AT THE HEAD OF THIS NOTICE.

UNCERTIFIED CHECKS WILL NOT BE ACCEPTED.

Defendant's Exhibit 4

SEP 25 '36 0[illegible] — [illegible]

PETITION

102613

TO THE COMMISSIONER OF PATENTS:

Sir:

Your petitioner THEODORE EMILE LEFORT, a citizen of the Republic of France and a resident of Paris in the Department of Seine, whose post office address is 8 Avenue Percier, prays that he may be allowed to surrender the Letters Patent for improvement in PROCESSES FOR THE PRODUCTION OF ETHYLENE OXIDE, No. 1,998,878, granted April 23, 1935, on an application filed by him on March 22, 1932, Serial No. 600,578, whereof Carbide and Carbon Chemicals Corporation, on whose behalf and with whose assent this application is made, is now sole owner, and that Letters Patent may be reissued to said Carbide and Carbon Chemicals Corporation for the same invention upon the annexed amended specification. With this petition is forwarded a request for an abstract of title duly certified to be filed with this petition, as required in such cases,

And he hereby appoints CHARLES C. SCHEFFLER, Register No. 11032, and EUGENE L. GREENWALD, Register No. 3276, both of 30 East 42nd Street, New York, New York, his attorneys with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to receive the Letters Patent, and to transact all business in the United States Patent Office connected therewith, the said power to be exercised separately or collectively by said attorneys as they shall from time to time elect. Please address all communications to Charles C. Scheffler.

Signed at Paris, France, this tenth day of September, 1936. Theodore Emile Lefort

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The undersigned, assignee of the entire right, title and interest in and to the above-mentioned Letters Patent, hereby assents to the accompanying application.

CARBIDE AND CARBON CHEMICALS CORPORATION

By _____
Vice-President

ATTEST

_____
Secretary

C 111

SPECIFICATION

TO WHOM IT MAY CONCERN:

Be it known that I, THEODORE EMILE LEFORT, a citizen of the Republic of France, residing at 8 Avenue Percier in the City of Paris and Department of Seine, have invented certain new and useful improvements in PROCESS FOR THE PRODUCTION OF ETHYLENE OXIDE, of which the following is a full, clear and exact description, such as will enable others skilled in the art to which it pertains to make and use the same,

IN THE UNITED STATES PATENT OFFICE

SEP 25 1936
U.S. PAT. OFFICE

Honorable Commissioner of Patents,
Washington, D. C.

Sir:

There are transmitted herewith Petition, Specification, and Oath relating to an application for re-issue Letters Patent No. 1,998,878 for improvements in "Processes for the Production of Ethylene Oxide", granted April 23, 1935, to Societe Anonyme; Societe Francaise de Catalyse Generalisee as assignee of Theodore Emile Lefort, and since assigned to Carbide and Carbon Chemicals Corporation.

There is also forwarded herewith a request that a certified copy of the abstract of title of the patent identified in the preceding paragraph be prepared for placement in the file of this application, as required in such cases. We also enclose check No. 208,020 for thirty dollars ($30.00) in payment of the Government fee in re-issue cases.

Annexed hereto are duly certified copies of French Patent No. 729,952, together with Patents of Addition thereto Nos. 41,255; 41,474; 41,484; and 41,810; and French Patent No. 739,562, together with Patent of Addition thereto No.41,811, which Patents and Patents of Addition are those referred to, and identified by the filing dates thereof, in the oath accompanying the original application, Serial No. 600,578, on which was granted United States Patent No. 1,998,878.

The original grant of Letters Patent No. 1,998,878 is not presented at this time, but will be forwarded for surrender at such time as notification is given that the application is otherwise in condition for allowance.

Respectfully submitted,

E. L. Greenwald
Attorney for Applicant

New York, N. Y.
September 23, 1936

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F/K

This invention has for its object a process for the production of ethylene oxide which consists essentially in subjecting ethylene to the action of oxygen of the air in the presence of suitable catalysts at temperatures between about 150° C. and about 400° C. If desired, water or hydrogen or both may be introduced for the purpose of facilitating or accelerating the reaction as more fully hereinafter described.

The main reaction involved in this process may be indicated as:

$$2\ CH_2{:}CH_2 + O_2 = 2\ CH_2.CH_2 \quad (O)$$

This reaction is, so far as I am aware, entirely novel, and the process of this invention for the first time provides a means for making ethylene oxide by the direct oxidation of ethylene, that is, employing molecular oxygen rather than atomic oxygen supplied chemically.

The reaction of my invention takes place at atmospheric pressure, but, if it is commercially desirable, superatmospheric pressures of, say, about 700 pounds per square inch, can be employed. I have also found that the yield of ethylene oxide can be somewhat increased by operating the process at pressures lower than atmospheric pressure.

As catalysts, I prefer to use those comprising or composed of silver, preferably in a form presenting an extended surface, such as silver powder or silver wire gauze. Other catalytic agents may be selected from the following:

(1) A mixture of antimony, bismuth, silver, lead, arsenic, nickel and tin, the proportions of which may vary;

(2) Silver activated by the incorporation therewith of minor proportions of gold, copper or iron;

(3) Gold, copper or iron, alone, or combined in various proportions;

(4) Bismuth and antimony alone;

(5) The following mixtures, in which the constituents may be present in any desired proportions:

Silver, gold, copper and iron,
Silver, gold and copper,
Silver, and gold,
Silver, copper and iron,
Silver, and copper,
Silver, and iron,
Copper, and iron;

(6) Mixture of antimony, lead, nickel and silver;

(7) Mixtures of bismuth, nickel, silver and lead;

(8) Mixtures of antimony, bismuth and silver or nickel; and

(9) Mixtures of bismuth and silver or nickel.

Generally speaking, suitable catalysts can be obtained from the metals: silver, antimony, lead, bismuth, nickel, tin, arsenic, gold, copper and iron, taken in all of their various combinations and in varying proportions. These metals may be used as such, as the oxides, or in various combinations of the metals with the oxides.

The ethylene can be that obtained from any convenient source, such as coke oven gas, hydrogenation of acetylene, dehydration of ethyl alcohol, and the like.

The oxygen used in the process can be oxygen of the air, and it is usually convenient to introduce oxygen by admitting air to the reaction.

Carbon dioxide may be formed in the process by secondary reaction, and the efficiency of the process can be increased by diminishing the tendency toward this secondary oxidation. I have found that the reaction giving carbon dioxide is, contrary to expectations, independent of that giving ethylene oxide, and probably proceeds according to the equation:

$$C_2H_4 + 3O_2 = 2H_2O + 2CO_2.$$

Thus, by introducing water, or a suitable volume of carbon dioxide, or both, with the reaction gases, this reaction is checked. The retarding effect on the total oxidation of ethylene to water and carbon dioxide is probably due to the partial pressures of the water or carbon dioxide added. In some cases, the addition of both water and carbon dioxide completely suppresses the total oxidation of ethylene, resulting in a marked increase in the efficiency of the process. The water, if it is used, may be admitted to the reaction vessel either in the liquid state, or as steam.

My experiments have shown that silver used as a catalyst in this process can be activated by introducing with it, small quantities of gold, or of copper, or of iron. A mixture containing silver, gold, copper and iron gives very satisfactory results, and allows a specific reaction; the only by-product being carbon dioxide which, of course, may be readily separated from the ethylene oxide. Only traces of aldehydes are formed.

Several methods of operation for carrying the process into practice will now be given by way of example only.

I

In suitable apparatus designed to resist high pressures, ethylene and air are separately compressed by means of appropriate compressors. The ethylene and air may also be compressed together, provided adequate precautions are observed to avoid explosion of the mixture.

These two compressed gases are then led together into a reaction tube capable of withstanding high pressures, and which contains a catalyst selected from those mentioned above. The catalyst-containing reaction tube is heated, for instance, to a temperature of 200°C. The reaction tube also communicates with a circulating pump which will permit the introduction of water under pressure.

The reaction will take place between the ethylene and oxygen of the air, and by controlling the flow of materials, the process may operate continuously. The ethylene oxide formed may be separated from the other gases, for instance, by condensation.

II

Silver, in the form of powder or wire gauze, with or without a support or carrier, is introduced into a suitable tube, such as one formed of silica. The tube and catalyst are then heated to between about 150°and 400° C., and a mixture of ethylene, air, water and hydrogen is passed through the tube. The volume of hydrogen used is about 5% of the total mixture.

Ethylene oxide is formed, and can be separated from the gas mixture issuing from the tube.

III

A silver colloid is placed in a high pressure tube which is filled with water. Pure ethylene under pressure is then added to form a solution of ethylene in the water.

The tube is thereafter heated between 150° C. and 400° C., and air and hydrogen are slowly introduced. The addition of air is stopped when all of the ethylene has been oxidized.

The ethylene oxide produced according to the process of this invention can be reacted with water in known manner to yield ethylene glycol, and, by other known methods, it can be converted into various derivatives of ethylene glycol. The reaction by which ethylene glycol is made may be indicated by the equation:

$$C_2H_4O + H_2O = CH_2OH.CH_2OH.$$

My experiments have shown that in the presence of the various catalytic materials indicated above, water, in the form of steam or not, considerably promotes the reaction insuring the production of ethylene oxide.

It will be clear that the process can be varied without departing from the essential teachings, and such modifications are included within my invention as defined by the appended claims.

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I claim:

1. A process for the production of ethylene oxide which comprises subjecting ethylene to the action of oxygen in the presence of a catalyst.

2. A process for the production of ethylene oxide which comprises subjecting ethylene to the action of molecular oxygen in the presence of a catalyst.

3. A process for the production of ethylene oxide which comprises subjecting ethylene to the action of oxygen in the presence of a catalyst at a temperature between about 150° C. and about 400° C.

4. A process for the production of ethylene oxide which comprises subjecting ethylene to the action of oxygen in the presence of a catalyst essentially containing silver.

5. A process for the production of ethylene oxide which comprises subjecting ethylene to the action of oxygen in the presence of a catalyst essentially containing silver activated by the addition of a small proportion of another metal.

6. A process for the production of ethylene oxide which comprises subjecting ethylene to the action of molecular oxygen in the presence of a catalyst essentially containing silver.

7. A process for the production of ethylene oxide which comprises subjecting ethylene to the action of oxygen at a temperature between about 150° C. and about 400° C. in the presence of a catalyst essentially containing silver.

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8. A process for the production of ethylene oxide which comprises subjecting ethylene to the action of oxygen of the air at a temperature between about 150° C. and about 400° C. in the presence of a catalyst essentially containing silver.

9. A process for the production of ethylene oxide which comprises subjecting ethylene to the simultaneous action of oxygen and water in the presence of a catalyst.

10. A process for the production of ethylene oxide which comprises subjecting ethylene to the simultaneous action of oxygen and water in the presence of a catalyst at a temperature between about 150° C. and about 400° C.

11. A process for the production of ethylene oxide which comprises subjecting ethylene to the simultaneous action of oxygen and water in the presence of a catalyst essentially containing silver.

12. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen end water at a temperature between 150° and 400° C. in the presence of a catalyst composed of silver activated by small quantities of a metal selected from a group consisting of gold, copper and iron.

13. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of the oxygen and water at a temperature between 150° and 400° C. in the presence of a catalyst composed of a mixture of silver, gold, copper and iron.

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3 ~~14.~~ A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water at a temperature between 150° C. and 400°C in the presence of a catalyst composed of a mixture of silver and gold.

4 ~~15.~~ A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen and steam at a temperature between 150° and 400° C., and at a pressure below atmospheric pressure, in the presence of a catalyst composed of a metal selected from a group consisting of silver, gold, copper and iron, and in the presence of hydrogen in the proportion of 5 per cent of the total volume.

5 ~~16.~~ A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen and steam at a pressure below atmospheric pressure, in the presence of a catalyst composed of a metal selected from a group consisting of silver, gold, copper and iron, and in the presence of hydrogen in the proportion of 5 per cent of the total volume, and of CO_2.

6 17. A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen at a temperature between 150° and 400° C. in the presence of a catalyst composed of silver activated with small quantities of gold.

7 18. A process for the production of ethylene oxide consisting in subjecting ethylene to the simultaneous action of oxygen, steam and hydrogen at a temperature between 150° and 400° C. in the presence of a catalyst composed of silver activated with small quantities of a mixture of gold and copper

C 120

IN TESTIMONY WHEREOF I AFFIX MY SIGNATURE. Theodore Emile Lefort

OATH

Republic of France :

Department of Seine : SS.

City of Paris :

REPUBLIC OF FRANCE, CITY OF PARIS } ss
CONSULATE GENERAL OF THE UNITED STATES OF AMERICA }

THEODORE EMILE LEFORT, the above-named petitioner, being duly sworn, deposes and says that he does verily believe himself to be the original, first and sole inventor of the improvement set forth and claimed in the foregoing specification and for which improvement he solicits a patent; that he does not know and does not believe that said improvement was ever before known or used; that he is a citizen of the Republic of France and resides at Paris in the Department of Seine; that he verily believes that the Letters Patent referred to in the foregoing Petition and Specification, and the surrender of which is hereby tendered, are in part inoperative and/or invalid for the reasons that the specification may be in part uncertain in describing the invention, and that there may be claimed therein as his invention and discovery more than he had a right to claim as new, and that there is failed to be claimed therein more specifically subject-matter of his invention which he had a right to claim as new, and that such defects consist particularly in the statements relative to the invention occurring at page 1, first two paragraphs, and in the claims, which fail to point out the particular scope of applicant's invention wherein ethylene is catalytically oxidized to ethylene oxide, and particularly by means of molecular oxygen.

Deponent further says that the errors which render such patent so inoperative and/or invalid arose from inadvertence, accident or mistake and without any fraudulent or deceptive intention on the part of deponent, and that the following is a true specification of the errors which it is claimed constitute such inadvertence, accident or mistake, to wit;

(a) The specification and claims do not emphasize sufficiently clearly that the process of oxidizing ethylene to ethylene oxide is operable whether or not water is present, and that the presence of water in the reaction is but one preferred embodiment of the invention; that this is the intent and contemplation is shown by the equation at lines 11 to 15, page 1, 1st column; at lines 34 to 44, page 1, 2nd column, concerning the probable effect of the added water; at lines 1 to 5, page 2, 2nd column, where it is stated that "***water, in the form of steam or not, considerably promotes the reaction***"; by lines 17 to 19, page 2, 1st column, where it is stated that means merely "allowing" the introduction of water are provided; and as will more fully appear by reference to the description of the process contained in the applications filed in France by deponent and referred to in the oath accompanying the original application.

(b) The specification and claims do not emphasize sufficiently clearly that silver, alone or activated, is the preferred catalytic agent for conducting the process of the invention; that this is the intent and contemplation is shown by reference to the second and third examples of the process at lines 28 to 30 and line 41, page 2, 1st column, and at lines 22 to 27, page 1, 2nd column.

(c) The claims are phrased in uncertain language and the grammatical construction thereof does not permit accurate definition of the invention, and there are failed to be claimed specific features of the invention which deponent had a right to claim as new, and which are claimed in claims 1 to 11, inclusive.

Deponent further says that such errors so particularly specified arose or occurred as follows: through failure of the applicant and his attorney to appreciate fully the relation of the claims to the preferred scope of the invention, and failure of applicant to instruct adequately his attorney in the fullest benefits and application of his invention, namely, in the production of ethylene oxide by oxidation of ethylene in the presence of catalysts with or without the inclusion of water in the reaction. This failure to appreciate fully these features of applicant's process arose, at least in part, through the fact that applicant is a resident of France and is not familiar with the law, rules and procedures relative to the application for and grant of United States Letters Patent coupled with the fact that applicant's attorney had no knowledge of or information relative to the invention other than that contained in the corresponding applications for patent filed in France. Recently the present assignee of the said Letters Patent, through its technical representatives and patent counsel, carefully studied applicant's process and the said Letters Patent, and observed and ascertained the defects and errors specified above, and the fact that the claims are uncertain and/or of breadth incommensurate with the invention. Promptly upon the discovery of the failure of the patent to include a clear statement of the invention as appears at lines 1 to 25 of page 2 of the present application, and to include claims in the form of those numbered 1 to 11, inclusive, of

the present application, this application for the reissue of the said Letters Patent No. 1,998,878 was prepared, and is submitted herewith.

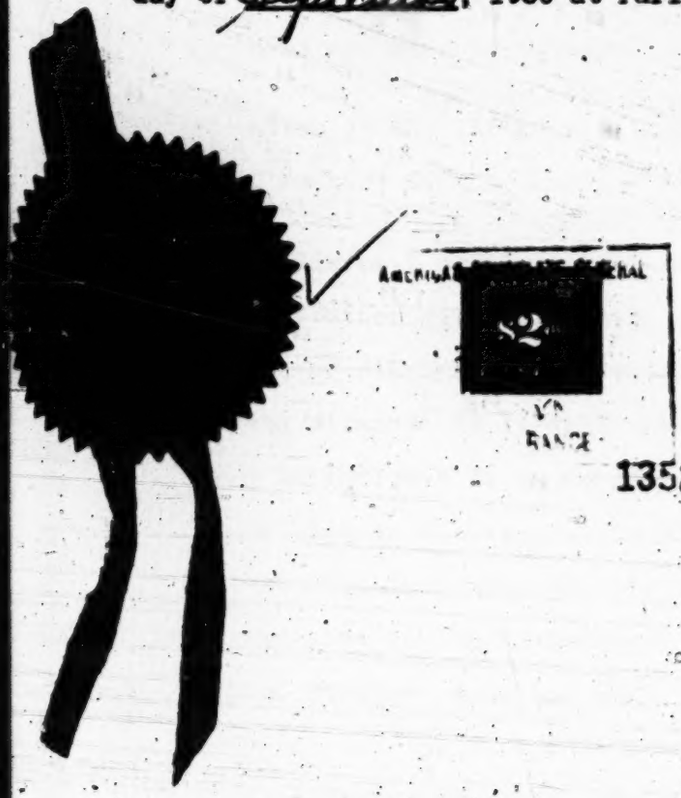
Theodore Emile Lefort

Sworn to and subscribed before me this tenth day of September, 1936 at Paris, France.

Paul C. Betts

United States Consul.

PAUL C. BETTS
Vice Consul of the United States of America at Paris, France



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Defendant's Exhibit 4 (Continued)

Div. 39 | Room 4709 ENG:EH

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
WASHINGTON

Paper No. 2

Please find below a communication from the EXAMINER in charge of this application.

Commissioner of Patents.

Applicant: T. E. Lefort

Ser. No. 102,613
Filed Sept. 25, 1936
For Process for the Production of Ethylene Oxide.

Charles C. Scheffler,
30 E. 42nd St.,
New York, N. Y.

MAILED
OCT 21 1936

This application has been examined and the following references are made of record:

Walker 972,953 Oct. 18, 1910 260/54
Willstatter et al, Liebig's Annalen der Chemie Vol. 422 (1920) pp. 36 to 46.
Lenher, Journal of the American Chemical Society, Vol. 54, 1932, pp. 1830-1837.

Claims 1 to 11 are rejected as without patentable merit over Willstatter cited herewith, who discloses inter alia the oxidation of ethylene in the presence of a variety of catalysts including silver. It is said that ethylene oxide may be attained by such oxidation. It is noted that in presenting the original case and also in distinguishing over this reference in the original applicant finally restricted the claims to specified catalyst and indicated that such catalysts and the presence of steam enabled increased yields. In other words, the case was presented as an improvement over prior known processes of oxidation of ethylene, so as to increase the yield of ethylene oxide. It appears to be too late to now urge that applicant is the pioneer inventor of processes of oxidizing ethylene.

It is noted that claims 9 and 10 now presented are virtually the same as claims 1 and 5 of the original, which were withdrawn after the citation of art. Claims 1 to 8 and 11 are of still broader scope and would accordingly be subject to the same rejection. A reissue proceeding is a proceeding only to correct inadvertent errors and not to recapture a broad field that was abandoned or dedicated after the citation of art.

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S.N. 102,015 - 2 -

Claims 1 to 11 are accordingly further rejected as not proper subject matter for reissue.

Claims 12 to 18 may be allowed as at present advised.

The original must be surrendered before allowance.

BB

Ott Shepard

Examiner.

description or disclosure of anything related to the Lefort process. The official letter, speaking of Willstätter, states: "It is said that ethylene oxide may be attained by such oxidation." It is not clear where this is said. Certainly it was never said by Willstätter nor by anyone acquainted with his work. Because of the preeminent position occupied by Willstätter in the field of organic chemistry, it is unquestionably true that his work reported in the Annalen was a greater deterrent to attempts to oxidize ethylene than was anything else in this art. Surely the fact that Willstätter was so completely lacking in success in the catalytic oxidation of ethylene to useful products is proof to all conversant with his work that this field would not be fruitful.

One of the affidavits submitted herewith is concerned specifically with the chemical history and chemical action involved in the oxidation of olefines, in particular ethylene. This affidavit shows the vast chemical distinctions between various types of oxidation carried out by prior workers including Willstätter, and the unique chemical action of the present invention by which Lefort succeeded in adding directly a single atom of molecular oxygen to the ethylene molecule. Dr. George O. Curme, Jr. occupies a position in aliphatic chemistry almost without parallel. He is regarded as the founder of a truly American chemical industry in the synthesis of aliphatic compounds from ethylene and its homologues, and is acknowledged to be one of the world's foremost organic chemists. He has wide acquaintance and personal experience with the subject matter of the Willstätter article and with the production of ethylene oxide. He, like Willstätter and following Willstätter, carried out researches on the oxidation of

U. S. PATENT OFFICE
APR 12 1937
DIVISION 59

IN THE UNITED STATES PATENT OFFICE

Div. 59, Room 4709 : Serial No. 102,613

T. E. Lefort : Filed Sept. 25, 1936

PROCESSES FOR THE PRODUCTION OF ETHYLENE OXIDE

Hon. Commissioner of Patents

Washington, D. C.

Sir:

In response to the Official Action of Oct. 21, 1936, there are submitted herewith an amendment, together with a supplemental oath by the inventor, and three affidavits, which will be presently discussed. As is pointed out in the remarks accompanying the amendment, the description of the invention has been revised extensively (in words, not in substance) to place it in better form, and this has been accomplished by providing a clean copy of the amended description to avoid confusion which probably would otherwise result. This action was approved by the Examiner at a conference March 13, 1937.

Earlier submission of the amendments and accompanying affidavits has been impossible since those signed in France travelled via the Queen Mary, which arrived April 5, 1937.

Claims 1 to 11 were rejected "as without patentable merit over Willstätter", whose article with Max Bommer on oxidation of ethylene with molecular oxygen appears at pages 36 to 46, Volume 422, Annalen der Chemie. This rejection will not apply to claims 19 and 20 which now replace the claims 1 to 11 cancelled without prejudice. Willstätter provides no

ethylene with molecular oxygen of the air, and learned nothing in any way suggestive of the formation of the product which Lefort now has shown how to make. It is believed that consideration of Dr. Curme's affidavit and the facts contained therein will so clearly remove the last vestige of doubt as to the relation of the Lefort process to such art as the Annalen article by Willstätter and Bommer as to render further comment superfluous.

The Examiner has noted in the Official letter that in presenting the original case and in distinguishing over the Willstätter article which was cited against the original claims, the claims were restricted to a specified catalyst indicated to be useful to increase yields of ethylene oxide. This occurrence was due to the double mistake of applicant's solicitor in the original application and of the Patent Office. The events leading to the misapprehension of the real nature of this invention by applicant's original solicitor will clearly appear from the facts stated in the supplemental oath of the inventor and the affidavits by his French attorney, M. Guichard, and the attorney in charge of the application in this country, Mr. Jakobsson. This unfortunate chain of circumstance, in which the inadvertence of the French solicitor and the inventor combined with the mistaken application by the Patent Office of art which in fact was inappropriate and inapplicable to the claims, clearly creates such a situation as the reissue statutes are designed to correct. The Supreme Court in its decision in Topliff v. Topliff, 145 U. S. 156, at page 171, succinctly summarized what should be done in such cases. The case of In re Heroult, 1907 C. D. 521 (App. D. C.) likewise discusses a remarkably similar situation in which a French inventor was inadvertently deprived of protection for

his real invention and was permitted to remedy that mistake by reissue. The remedy sought in this case is no more than equity demands and precedent sanctions.

The Examiner has pointed out that the original application was presented as an improvement over prior known processes of oxidising ethylene. Applicant is perfectly willing to have the invention regarded as an improvement on previous processes of oxidizing ethylene by means of molecular oxygen, so long as it is clearly understood that the "improvement" provided by him is that of for the first time making possible the addition of a single atom of oxygen from molecular oxygen to the ethylene molecule to give for the first time ethylene oxide as a direct product of this oxidation. It is not too late to acknowledge the fact that applicant has made a basic invention and has pioneered an entirely new type reaction in organic chemistry which has the outstanding distinctions of being both chemically unique and industrially valuable.

Certain of claims 1 to 11 were rejected as being of substantially the same, or of broader, scope than claims which were cancelled from the original application after the citation of art. Specifically, the Examiner referred to the cancellation of claims 1 to 5, which were rejected in the original application by the Official letter dated June 24, 1933 on reference to the Willstätter article, supra, in view of an article by Lenher, J.A.C.S. 53, pages 2420 to 2421; 1931. Following this rejection, claims 1 and 5 were cancelled by an amendment filed December 22, 1933. It is clear that both the rejection of those claims in the original application and their cancellation were due to the inadvertent mutual mistake on the part of the

Patent Office and applicant's then solicitor. The Patent Office erred in applying the article by Lenher to the rejection of claims both by reason of its lack of relevancy to the process claimed by Lefort and because its publication date, namely June 8, 1931, was subsequent to the date of March 27, 1931, available to the applicant under the provisions of the International Convention. In similar fashion, applicant's solicitor inadvertently failed to point out these two facts to the Examiner and mistakenly acquiesced in the position of the Office. Since the withdrawal of the claims 1 and 5 from the original application was due to a mistake of fact and not to an error in judgment on the part of applicant or his solicitor which could have been remedied by appeal, it is clear that there is no estoppel against the present claims which can arise from this action. Moreover, claims 19 and 20, which have replaced claims 1 to 11, are not open to this ground of rejection even if it were in fact valid,- which it is not. The claims 19 and 20 which are now sought to protect applicant's invention have been carefully drafted to include all of the essential features of the Lefort process, and have been restricted in number for the convenience of the Office and to simplify prosecution of this application. These claims much more accurately define the invention than those which they supplant, and applicant is willing to rely on them for the protection of his invention, and chooses not to press further for other claims to his invention. Because these claims include as elements of the process certain features which were not heretofore contained in any claims drafted to this invention, it is plain that they do not represent any attempt to retrieve anything which could have been abandoned or lost to the public. The allowance of these

47. claims is believed to be clearly in order, and the facts

justifying this will be apparent from the oath of the inventor and the affidavits which have been submitted. There is no existing reason for not permitting applicant to secure protection for his invention in scope defined by claims 19 and 20, and this is especially the case where, as here, the invention is basic. Authority for all of the foregoing contentions will appear from the digest contained in Section 274, pages 354 and 355, "Walker on Patents", 6th Edition, Volume I.

Consideration of statements in the oath of the inventor annexed to the amendments to the description and claims, together with the accompanying affidavits of Dr. Curme, M. Guichard and Mr. Jakobsson, is urged. It is asked that the application be passed to issue in its present form, and the original patent will be surrendered as soon as such action is indicated.

Respectfully submitted,

Eugene A. Greenwald
Attorney for Applicant

New York, N. Y.
April 7, 1937

F/D

U. S. PATENT OFFICE
APR 19 1937
DIVISION

APR 10 1937
PATENT OFFICE

IN THE UNITED STATES PATENT OFFICE

Div. 59, Room 4709 : Serial No. 102,613

Theodore E. Lefort : Filed September 25, 1936

PROCESS FOR PRODUCING ETHYLENE OXIDE

Hon. Commissioner of Patents

Washington, D. C.

Sir:

In response to the Official letter of October 21, 1936, please amend the above application as follows:

Cancel pages 2 to 6, inclusive, of the specification, and rewrite as follows:

This invention provides a specific and novel process for making ethylene oxide. It essentially consists in causing ethylene to combine directly with molecular oxygen at temperatures of about 150° to about 400° C. in the presence of a surface catalyst which favors the oxidation of ethylene to ethylene oxide under these conditions. The process can be carried out at atmospheric pressure, and it may also be conducted at pressures above or below atmospheric pressure.

This process is characterized by an entirely unique reaction in which the direct addition of a single oxygen atom to the ethylene molecule occurs without the formation of significant amounts of other oxygenated compounds such as aldehydes. This novel reaction may be expressed as:

$2\ CH_2{=}CH_2 + O_2 \longrightarrow 2\ CH_2\text{-}CH_2$ (with O) and it is accompanied by complete oxidation of some of the ethylene to give carbon dioxide and water according to the equation:

$CH_2{=}CH_2 + 3\ O_2 \longrightarrow 2\ CO_2 + 2\ H_2O$

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Ethylene from any desired source may be used, including that obtained from coke oven gas, hydrogenation of acetylene, or dehydration of ethyl alcohol. The oxygen can be the oxygen of the air, and can conveniently be mixed with the ethylene prior to the reaction.

The oxidation of ethylene takes place with a giving off of heat, and it is, of course, desirable to maintain the temperature of the zone of reaction within the range specified. This can be facilitated by suitable dilution of the reaction gases, such as that accomplished by the use of air as the source of oxygen, and some water or carbon dioxide in addition to that formed can be admitted to the mixture in the reaction zone if desired. Hydrogen may be similarly added.

The process can be conveniently conducted by passing a mixture containing air and ethylene through a tube, for example, of silica or of a material capable of withstanding high pressures, within which is placed such a surface catalyst as one composed of silver in finely divided form, and which may be disposed within the tube on a supporting material, or in other known ways. The temperature of the reaction zone is maintained between about 150° and about 400° C., for example, at about 200° C., and the issuing products from the tube contain ethylene oxide, carbon dioxide and water without objectionable aldehydes. The ethylene oxide can be recovered in any suitable way, preferably in liquid form by condensation or solution.

The surface catalyst is preferably silver as stated, but other metals such as bismuth and antimony may be used. Likewise oxides of these metals or mixtures containing these metals or their oxides may be used. Experiments have also indicated that the catalyst may be silver activated by the addition of small amounts of gold or copper or iron.

In any case, the ethylene and oxygen are thus reacted simultaneously at the temperatures set forth in the presence of a surface catalyst and of water, and the process is conducted to produce ethylene oxide as a direct product in substantial and economic amount. The ethylene oxide can be recovered by known methods, and it may be further reacted to form ethylene glycol or derivatives of ethylene glycol.

Cancel claims 1 to 11, inclusive, without prejudice, and substitute the following:

-- ~~12.~~ The process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene, which comprises forming a mixture containing ethylene and molecular oxygen and conducting said mixture through a confined reaction zone which is maintained at an elevated temperature; controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400° C.; subjecting said mixture in said zone at said elevated controlled temperature to intimate contact with an active surface catalyst of a material having the catalytic characteristics of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water and which does not favor the existence in said products of substantial amounts of aldehydes; correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect

said oxidation of said ethylene directly to form substantial amounts of ethylene oxide; and thereafter recovering ethylene oxide-containing product in liquid form.

20. The process of making ethylene oxide by the direct chemical combination of oxygen with ethylene in the proportions of one atom of oxygen to one molecule of ethylene, which comprises forming a mixture containing ethylene and molecular oxygen and a heat-absorbing diluent and conducting said mixture through a confined reaction zone which is maintained at an elevated temperature; controlling said temperature to maintain said mixture in said zone at a temperature between about 150° and about 400° C.; subjecting said mixture at said elevated controlled temperature to intimate contact with an active surface catalyst material composed essentially of silver which favors the direct oxidation of said ethylene in said mixture by said molecular oxygen to form oxidation products containing ethylene oxide in the presence of water and which does not favor the existence in said products of substantial amounts of aldehydes; correlating the passage of said mixture through said reaction zone in the presence of said catalyst and the controlled elevated temperature of said mixture and said catalyst to effect said oxidation of said ethylene directly to form substantial amounts of ethylene oxide; and thereafter separating ethylene oxide from the products of oxidation.

REMARKS

In order to place this application in better form, applicant deems it essentially desirable to make numerous revisions in the description of his invention. Because these

revisions would result in extensive amendments to the specification now on file and lead to possible confusion, a completely rewritten description of the invention has been provided in clean copy. The rewritten description principally provides a representation of the disclosure of the invention from which portions of the original description have been deleted and the whole rephrased in clearer language. This matter was discussed at a conference with the Examiner on March 13, 1937, at which time informal approval of the above amendments containing the rewritten description was received.

The supplemental oath and the affidavits attached will make apparent the reasons for the several amendments, and will clearly show that claims 19 and 20, which replace the claims 1 to 11, hereby cancelled without prejudice, are allowable. Claims 12 to 18 have previously been allowed, and allowance of claims 19 and 20 is respectfully asked.

Respectfully submitted,

Eugene L. Greenwald
Attorney for Applicant

New York, N. Y.
April 7, 1937

F/D

SUPPLEMENTAL OATH

REPUBLIC OF FRANCE :
:
CITY OF PARIS :
: ss.:
CONSULATE GENERAL OF THE :
UNITED STATES OF AMERICA :

THEODORE EMILE LEFORT, whose application Serial No. 102,613, for Reissue of Letters Patent No. 1,998,878, was filed September 25, 1936, being first duly sworn, deposes and says that he does verily believe himself to be the original, first and sole inventor of the improvements in PROCESSES FOR THE PRODUCTION OF ETHYLENE OXIDE set forth and claimed in said application Serial No. 102,613 and forming the subject matter of the annexed amendment to said application and for which improvements he solicits a patent; that he does not know and does not believe that said improvements were ever known or used before his invention or discovery thereof, or patented or described in any country before his invention or discovery thereof or more than two years prior to his original application Serial No. 600,578 for a patent therefor, or in public use or on sale in the United States more than two years prior to his said original application for a patent therefor; that said invention has not been patented in any country foreign to the United States on an application filed by him or his legal representatives more than twelve months prior to his original application Serial No. 600,578; and that no application for patent on said improvement was filed by him, or his representatives or assigns in any country foreign to the United States, before the filing of his said application Serial No. 600,578, except as stated in his Oath

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accompanying his said original application; that he is a citizen of the Republic of France and resides in the City of Paris; that he verily believes that the Letters Patent No. 1,998,878 referred to in the Petition and Specification of said application Serial No. 102,613, and the surrender of which has been thereby tendered, are in fact inoperative and/or invalid for the reasons set forth in the oath annexed to and forming part of said application Serial No. 102,613, and for the reason that the specification is defective and insufficient and that such defects and insufficiencies consist particularly in the failure to adequately claim the real invention, in the failure to point out and explain the particular scope of deponent's invention which consists essentially in the oxidation of ethylene by means of molecular oxygen in the presence of silver or other suitable catalyst which acts in the process like silver to form ethylene oxide in substantial amount and without substantial amounts of aldehydes and then utilizing the ethylene oxide product, and in the innocent mis-description in the specification of several substances as satisfactory catalysts; and deponent further says that the errors which render such patent so inoperative or invalid arose from inadvertence, accident and mistake, and without any fraudulent or deceptive intention on the part of deponent; that the following is a further true specification of the errors which it is claimed constitute such inadvertence relied upon; the failure to present claims more commensurate with the real invention, the absence of claims in said patent which are operative to secure

to deponent his real invention, the failure more clearly to explain the particular scope of the real invention, the failure more clearly to point out that a substantial quantity of water is inherently present in the reaction tube due to the substantial independent oxidation of ethylene to carbon dioxide and water, the failure to make clearer that the addition of water or carbon dioxide or both to augment the water and carbon dioxide inherently and independently formed, as well as the addition of hydrogen, are merely supplemental features still within the scope of the real invention, and the description of certain substances as alternative catalysts which deponent now disclaims namely, lead, nickel, tin, arsenic, gold, copper and iron per se, and further in the failure of the specification to contain a description of the invention in the words and language provided by the annexed amendment to the specification of said application Serial No. 102,613, and in the failure to claim as deponent's invention or discovery that which he had a right to claim as new, and which is claimed in claims 19 and 20 of the annexed amendment to application Serial No. 102,613; and that such errors so particularly specified arose as follows:

Deponent is totally unfamiliar with the laws and rules governing the issuance and construction of Letters Patent of the United States. Until shortly before the application for reissue of the said patent, deponent was ignorant of the existence of any difference between French patent law and that of the United States. Not being advised of such difference during the prosecution of the application

resulting in the issuance of said Letters Patent or until shortly before the application for reissue thereof, deponent believed that the United States patent law was the same as that of France. Accordingly, deponent believed that, being the first to disclose the invention which is the subject of said Letters Patent, he thereby would and did secure, by the issuance of said Letters Patent, whatever might be new as therein described irrespective of the language of the claims. Deponent further believed that, as he was the first to disclose a process of oxidizing ethylene by the oxygen of the air and the aid of a catalyst into ethylene oxide, he obtained the fullest protection of his invention by the issuance of Letters Patent which merely set forth all specific features which he contemplated as original with him.

Deponent says that on March 27, 1931, when he applied for his first French patent on the said invention, and at all times thereafter, he verily believed that he was the first to discover and disclose that molecular oxygen could be made to react with ethylene in the presence of a surface catalyst to form ethylene oxide without aldehydes in substantial amounts.

Deponent further states that he failed to present adequate claims to his real invention because he believed that the protection afforded by the patent was delineated by the disclosure as in French law, and that the claims were, as in the case of the resumé in a French patent, a mere formal appendage of no controlling importance. A resumé in a French patent is in the nature of a formality and does not control the protection afforded by the disclosure of the

patent. Throughout the entire prosecution of the application deponent, due to these mistaken beliefs, centered his attention upon retaining a description of the invention in its narrower aspects of possible specific variations, relying on the range of disclosure rather than on the claims to afford scope of protection to which he believed, and still believes, himself entitled.

Deponent through inadvertence failed to stress to his attorney the point that a substantial quantity of water was inherently present by reason of independent complete oxidation of some of the ethylene giving water and carbon dioxide, and that any augmentation of this water by the direct addition of water to the reaction zone might be practiced or omitted as desired.

Deponent further states that at no time did he explain to his attorney, L. Guichard, the scope of the said invention with respect to the prior art, and that M. Guichard was the attorney of the firm of Elluin and Barnay who was solely responsible for all patent applications with respect todeponent's invention.

Deponent further says that he verily believes that his said attorney, through inadvertence and mistake, believed that the oxidation of ethylene to ethylene oxide comprised the formation of formaldehyde as an intermediate product. Deponent believes that his attorney, through inadvertence and mistake, believed that references disclosing methods for obtaining formaldehyde by the oxidation of ethylene anticipated the broad aspect of deponent's invention.

Deponent further says that these erroneous beliefs of his attorney arose by reason of deponent's inadvertence and mistake in failing to communicate his invention to his attorney by pointing out what was excluded from it as well as what was included in it. It was only after the issue of deponent's patent No. 1,998,878 that deponent discovered the defects thereof now sought to be corrected and discovered the matters of inadvertence, accident and mistake which combined to occasion them.

Deponent further says that he neglected adequately to impress his attorney to note that the date of the publication of an article by Lenher in Journal of the American Chemical Society, Volume 53, pages 2420-2421, namely, June 8, 1931, was later than the priority date of March 27, 1931 on which deponent relied, and to which deponent was entitled by virtue of the International Convention.

Deponent further says that the catalyst first used in practicing his invention was a commercial grade of antimony containing a substantial quantity of arsenic as an impurity therein. This mixture was an effective catalyst. Later a spectrographic analysis of this mixture was made and it was found that it was composed of the substances set forth at page 1, column 1, lines 23-5 of the patent. From this it was concluded first that none of these substances acted as negative catalysts. Later runs were made with chemically pure antimony and the results were inferior to those obtained with commercial antimony. From this it was concluded that these impurities were themselves catalysts. Then, before all these substances which were present in the

commercial antimony as impurities could be studied as to their individual catalytic effects, deponent found that silver was the most excellent catalyst for the reaction. After that time deponent directed efforts solely toward exploring the possibilities of the use of silver as a catalyst. In his application for United States patent, deponent endeavored to disclose as fully as possible the results of his work and permissible deductions therefrom. Therefore deponent included in his description all substances which he had reason to believe were suitable catalysts but endeavored at the same time to make clear that silver, antimony and bismuth were the principal catalysts with which he was concerned (patent p. 1, column 1, 11, 26, 27, 28, 31-2, 48-9, 51; p. 2, column 1, 11, 28-49).

Lest deponent's beliefs as to lead, nickel, tin, arsenic, gold, copper and iron per se be questioned on the ground that they were premised on insufficient evidence, deponent is willing to exclude mention of such metals from his specification and confine his disclosure to those metals used in successful tests.

Chevalier Emile Lefort

Sworn to before me this 30th day of March 1937, at Paris, France.

United States Consul
JOHN R. WOOD
Vice-Consul of the United States
of America at Paris, France

3082

DIVISION
APR 10 1937
U. S. PATENT OFFICE
U. S. PATENT OFFICE
APR 19 1937
DIVISION 59

IN THE UNITED STATES PATENT OFFICE

Div. 59, Room 4709 : Serial No. 102,613

Theodore E. Lefort : Filed Sept. 25, 1936

PROCESS FOR PRODUCING ETHYLENE OXIDE

A F F I D A V I T

State of New York :
: ss.
County of New York :

GEORGE O. CURME, JR., vice-president of Carbide and Carbon Chemicals Corporation, New York, New York, a citizen of the United States and a resident of White Plains, New York, being duly sworn, deposes and says:

1. QUALIFICATIONS

I am a chemist with the degrees of Bachelor of Science and of Doctor of Science (honorary) from Northwestern University and the degree of Doctor of Philosophy from the University of Chicago, and my graduate studies in chemistry additionally include study at Harvard University, at the Kaiser Wilhelm Institute and at the University of Berlin in Germany.

In 1933 the Chandler medal was presented to me by the Charles Frederick Chandler Foundation for research in synthetic aliphatic organic chemistry and for the advancement of industry through its commercial applications; in 1935 I received the Perkin medal, awarded by the American Section of the Society of Chemical Industry for advancement of applied

chemistry through developments with which I have been associated in bringing into commercial production new products derived from ethylene and propylene and including ethylene oxide, ethylene glycol, ethylene dichloride, ethylene chlorhydrin, ethyl alcohol, isopropanol and acetone; and in 1936 I received the Elliot Cresson medal, awarded by the Franklin Institute, Philadelphia, for research which resulted in the development of industry based on synthetic aliphatic compounds, derived from the olefines.

For the past twenty-three years I have been engaged in and intimately associated with research in aliphatic chemistry, and in the industrial development of synthetic aliphatic chemistry, and I have for the past seventeen years been respectively Chief Chemist, Director of Research and Vice-President of Carbide and Carbon Chemicals Corporation, which organization has been active in technical research and commercial development of new chemical products and processes derived from ethylene and the homologous olefine gases, prominent among which products have been ethylene glycol and other derivatives of ethylene oxide.

As a result of my studies, activities and interests in the fields set forth, I am thoroughly familiar with the technical and patent literature concerning particularly the production of ethylene oxide and ethylene glycol or derivatives of ethylene oxide from starting materials such as ethylene.

I am familiar with patent 1,998,878 granted April 23, 1935 to T. E. Lefort, and with the application Serial No. 102,613 for reissue of patent 1,998,878, which application was

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filed September 25, 1936, and I have been intimately associated with further research and development on the process for the production of ethylene oxide by the direct, catalytic addition of a single oxygen atom to ethylene which forms the subject matter of that patent and the application for its reissue.

2. HISTORY OF DIRECT OXIDATION OF OLEFINES

To the best of my knowledge and belief, the history of the direct oxidation of olefines is as follows:

Uncatalyzed Oxidation

The study of the reactions of olefines with molecular oxygen (O_2) dates back to 1879, when Schutzenberger discovered that the controlled oxidation of ethylene with the molecular oxygen of air at about 400° C. gives recognizable amounts of formaldehyde. Bone and Wheeler studied this reaction, mostly in the absence of catalysts, and found that it is quite slow below 500° C. They obtained only traces of formaldehyde, since this product is readily further oxidized to oxides of carbon and water.

Richard Willstätter and Max Bommer (Ann. 422, 36; 1920) studied the oxidation of ethylene with the molecular oxygen of air for the practical purpose of making formaldehyde. They found that, when the ethylene is dilute, the contact time short and the temperature about 600° C., it is possible to remove the formaldehyde from the reaction zone before it has been completely oxidized. However, their highest reported yield of formaldehyde represented only 3% of the ethylene introduced and 50% of the ethylene reacted. Willstätter patented the process (Ger. pat. 350,992), but apparently it had no

commercial success. Blair and Wheeler (J.S.C.I. 41, 303T; 1922) studied the same reaction at length, and in general confirmed the conclusions of Willstätter and Bommer.

Shortly after the publication of the above article by Willstätter and Bommer, experimental work on the oxidation of ethylene with molecular oxygen of the air was carried out under my personal direction in the laboratories of Carbide and Carbon Chemicals Corporation. The results obtained in this work, which was not published, were substantially the same as those reported by Willstätter and Bommer.

Higher olefines were found, like ethylene, to give aldehydes on oxidation with molecular oxygen. Engler (Ber. 33, 1094; 1900) and Lewis (J.C.S. 1929, 759) showed that in the low temperature oxidation of amylene with the molecular oxygen of air, peroxides are formed to a certain extent as intermediates. In general, peroxides are formed as the primary products of the oxidation of olefinic substances with molecular oxygen, as shown by the following equation:

$$H_2C = CH_2 + O_2 \longrightarrow H_2C\!-\!CH_2 \quad \text{(O–O)} \qquad \text{I.}$$

These are highly unstable substances which usually break down wholly or in part under the conditions of their formation to secondary products, such as aldehydes, olefine oxides, organic acids and oxides of carbon.

I believe the first intimation in the chemical literature that ethylene oxide can be made by the uncatalyzed oxidation of ethylene with molecular oxygen was published as a short "Communication to the Editor" by Lenher (J.A.C.S. 53, 2420; June 8, 1931). According to Lenher, the oxidation of

ethylene with the molecular oxygen of air seems to take place in two ways; one being the direct oxidation of ethylene to formaldehyde, and the other being the formation of a peroxide which in turn yields ethylene oxide, dioxymethyl peroxide, hydrogen peroxide, formaldehyde and other products. Since organic peroxides in general are highly unstable at elevated temperatures, it is not surprising that Lenher obtained a multiplicity of products, resulting from the decomposition of the primary peroxide, and only small yields of any one product. In order to produce ethylene oxide, Lenher used lower reaction temperatures than those of Willstätter and circulated the gases through the reaction zone and through an apparatus for absorbing the condensable reaction products. Only traces of ethylene oxide were formed in each passage through the reaction zone, but the eventual effect of an uncounted number of passes was to accumulate appreciable amounts of ethylene oxide in the absorber. The amount of ethylene consumed per pass was also small, but sufficient that at best the ethylene oxide obtained was equivalent to some 20% of the ethylene consumed. The low ultimate yield would indicate that commercial success of the process is unlikely, and the fact that only traces are produced per pass seems to exclude the process as described entirely from commercial consideration. A patent on the process (U. S. 1,995,991) was assigned to E. I. du Pont de Nemours & Company, but apparently no attempt has been made to operate it on a substantial scale.

Beside the low amount of reaction per pass, a distinguishing characteristic of Lenher's procedure was that wide, empty tubes or other reaction vessels were employed, whereas

Willstätter used tubes having a diameter of only 3 mm. Lenher found that the reaction giving ethylene oxide was prevented or hindered by the catalytic action of any surface such as those of narrow or packed tubes. He concluded that the reaction is a chain reaction occurring in the gas phase, and that the chains are shortened or prevented by surfaces in general. Such chain reactions, that is, reactions in which the energy necessary for activation is passed on from molecule to molecule, have in recent years been found to occur in many cases of oxidation.

Catalyzed Oxidation

Early in the work on the oxidation of olefines with molecular oxygen, it was recognized that catalysts accelerate such reactions. There is no report, however, of any successful catalytic oxidation involving only the direct addition of a single oxygen atom to the ethylene molecule. Acceleration of the oxidation reaction by metals and oxides was noted by Phillips (Z. anorg. Ch. 6, 213; 1894), Bone and Smith (J.C.S. 87, 910; 1905), and Sabatier (Die Katalyse in der organischen Chemie). Willstätter and Bommer reported that oxidation catalysts in general lower the temperature at which effective oxidation of ethylene with molecular oxygen takes place. They apparently studied this sort of oxidation quite thoroughly, since they had reason at the outset to suppose that catalysts would help them. The lower reaction temperature would be expected to be favorable, especially because some of the oxidation catalysts were known to be effective in producing formaldehyde by the oxidation of methanol. However, the results from their catalytic work were apparently unproductive, and the

work was not referred to in their patent, but mentioned briefly in their article in the Annalen (supra). The catalysts tried by these authors to promote the oxidation of ethylene with molecular oxygen did not yield formaldehyde or any other valuable product, so far as can be learned from their writings.

On the other hand, Wheeler and Blair found that under certain conditions the catalytic oxidation of ethylene with molecular oxygen yields some formaldehyde. Various processes have been proposed for making aldehydes and acids from olefines by the use of various oxidation catalysts. Quite generally, however, these processes involve substantial changes in the olefine molecule, usually splitting it into two parts at the double bond, as for example ethylene is believed to split when it is oxidized to formaldehyde. Lenher's work, referred to above, seemed quite convincing that the catalysts which he used are detrimental to the formation of oxidation products of ethylene retaining the original carbon to carbon linkage just as had been found by Willstätter. The following statement was made in a textbook by L. F. Hareck, Acting Director of the Research Laboratory of Applied Chemistry, Massachusetts Institute of Technology, and D. A. Hahn, Professor of Organic Chemistry, Mount Holyoke College (The Catalytic Oxidation of Organic Compounds in the Vapor Phase, 1932, p. 212): "The direct applicability of catalysts to such an oxidation reaction (oxidation of ethylene to its oxide) for the purpose of increasing rate of reaction or increasing conversion to ethylene oxide is highly questionable in the light of present knowledge regarding chain reactions."

The first intimation in the literature that ethylene glycol, a substance related to ethylene oxide, can be made by

the oxidation of ethylene using iodine as a catalyst was published by Skärblom (U. S. 1,982,545). His process does not depend on surface catalysis, like the experiments mentioned above, but is a combination of a series of well-known reactions in the liquid phase, in homogeneous aqueous solution and at temperatures below the boiling point of water. In this reaction the ethylene reacts with iodine and with hypoiodous acid, forming ethylene diiodide and ethylene iodohydrin. These are hydrolyzed, giving ethylene glycol and hydriodic acid. The hydriodic acid finally reacts with free oxygen, regenerating iodine. The process has not been found commercially successful and need not be further discussed, since it is obviously very different from a surface catalysis of the oxidation of ethylene.

The direct addition of a single oxygen atom to an olefine bond by surface catalysis was discovered by T. E. Lefort (F. P. 729,952, March 27, 1931). His process is distinguished from Willstätter's formaldehyde process in that Lefort used certain catalysts, and obtained substantial yields of ethylene oxide, but no recognizable amounts of formaldehyde or other reaction products, excepting only carbon dioxide, water and a trace of acetaldehyde. On the other hand, Willstätter used the uncatalyzed oxidation to produce formaldehyde but no recognizable amounts of ethylene oxide.

Viewed from present knowledge, Lefort's process might be academically argued to have something in common with the laboratory experiments which Willstätter made, using various catalysts. Such a comparison could be made only by hindsight and only by one intentionally blind to the difference

between success and failure. Willstätter mentioned or implied in his paper with Bommer (Ann. 422, 36; 1920) that he made such experiments, but he did not describe them in this paper or in any other publication known to me. Apparently Willstätter and Bommer tried unfruitfully two of the same catalysts, copper and silver, which Lefort later found useful to give ethylene oxide. The net result of this experimentation by Willstätter and Bommer added nothing to the store of useful human knowledge regarding the direct addition of a single oxygen atom to ethylene which Lefort subsequently showed how to conduct easily and with excellent yield.

Richard Willstätter is widely recognized as one of the greatest organic chemists of the present time. He made great advances in extremely difficult researches, such as the studies of chlorophyll, carotenoids, and enzymes. Among many other honors, he received the Nobel Prize for chemistry in 1915, and the Willard Gibbs Medal at Chicago in 1933. It might seem obvious that a chemist of Willstätter's caliber would recognize the possibility of forming ethylene oxide and the importance of testing for it. If so, he evidently did not find any. In his article in the Annalen (supra) he described his experiments without catalysts, merely mentioning those with catalysts without a description, and in the corresponding patent (Ger. 350,922) he does not even mention specifically his catalytic experiments, but classes them along with those of others, as resultless.

Comparison of Catalyzed and Uncatalyzed Oxidation

Lefort's process is easily distinguished from Lenher's in that Lefort's depends on the surface action of certain catalysts, whereas Lenher's is inhibited or entirely prevented

by the action of surfaces. Chemically Lefort's reaction is unique, since it adds directly one atom of oxygen from molecular oxygen (O_2) to the olefine linkage, forming ethylene oxide, in accordance with the following equation:

$$H_2C = CH_2 + O \longrightarrow H_2C\!-\!CH_2 \quad \text{(O bridging)} \qquad \text{II.}$$

In a wholly different manner, Lenher's reaction adds both atoms of a single oxygen molecule to the olefine linkage of ethylene, forming ethylene peroxide, which then breaks down giving various products (see Reaction I.). The formation of peroxides from olefine bonds by the direct addition of two atoms (i.e. a molecule) of oxygen has long been known, but Lefort's reaction, starting with free molecular oxygen and adding one atom only to the double bond, is chemically novel. It seems likely that Lefort's reaction is applicable quite generally to other olefines as well as to ethylene. In fact, its applicability to propylene has already been reported. However, the field opened by this discovery is so new that many years of study may be required before its real scope can be ascertained.

From the point of view of practical interest, Lefort's process is distinguished from Lenher's in that the yields are greatly superior. Not only does Lefort's primary reaction tend to go several times higher in the direction of ethylene oxide than that reported by Lenher for his reaction, but the conditions of operation of Lefort's reaction are such that ethylene oxide is relatively stable, as compared with the conditions of low stability reported by Lenher for his reaction. For this reason, in Lefort's process it is not necessary to limit the yields to traces of ethylene oxide per pass

through the reaction zone, but as much as 45% of the ethylene can be usefully converted to ethylene oxide in a single pass. This difference alone is sufficient to distinguish between commercial practicability and impossibility, because of the enormous amount of gases which would have to be handled in putting the process described by Lenher on a commercial scale.

In contrast to Lenher's process, which always makes more formaldehyde than ethylene oxide, Lefort's process may give total aldehydic impurities amounting to not more than 0.2% of the ethylene oxide produced. This aldehydic impurity appears to be principally acetaldehyde and contains formaldehyde in minute proportion, if at all. The acetaldehyde content of 0.2% is less than half of that contained in ethylene oxide of refined commercial grade, made by the current chlorhydrin-ethylene oxide process, now operating on a large industrial scale.

Ethylene oxide and the allied substances, ethylene glycol and its ethers, have become important commercial products in the past ten or fifteen years. The known practical processes for making these materials involve the reaction of ethylene, chlorine, and water to give ethylene chlorhydrin. About two pounds of chlorine are required per pound of ethylene oxide, and this chlorine is finally eliminated in the form of by-products. The commercial importance of a practical direct oxidation process, so far provided only by Lefort, is thus obvious.

3. TECHNICAL ASPECTS OF LEFORT'S PROCESS

During the past few months, the process disclosed in Lefort's patent and the application for reissue has been

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intensively investigated in the laboratories of Carbide and Carbon Chemicals Corporation under my direction, and I am familiar with that work and the results thereof stated below are accurate within reasonable limits of experimental error:

The catalytic oxidation of ethylene to produce ethylene oxide as a direct product proceeds satisfactorily with the use of silver or promoted silver catalysts, and catalysts of these types have been found by test to be preferable for the conduct of the process. The ethylene oxide is formed in substantial and economically valuable amounts. Tests have shown that overall yields of ethylene oxide, based on a single pass of the gases over the catalysts, as great as 45% can be consistently obtained. The efficiency of this reaction, crediting the ethylene unattacked during the first pass, is consistently above 60%, and under preferred conditions is as high as 80%. The oxidation proceeds to give results similar to those above stated at temperatures as low as 150° C. and as high as 400° C. Naturally there are certain variations in the overall yield and chemical efficiency throughout this temperature range, and the optimum temperature fluctuates within this range depending upon the related conditions of gas velocity, concentrations of ethylene and oxygen employed, and amount of active catalytic surface used. The chemical characteristics of this reaction are substantially unaffected by variations in pressure ranging upward from atmospheric to 100 psi.

Tests have shown that the proportions of ethylene and oxygen used have little, if any, effect on the chemical characteristics of the reaction. Ethylene concentrations from

1% to 80% have been tested, and oxygen concentrations from 2% to 20% have been employed. Mixtures of oxygen and nitrogen from any source may be used for reasons of economy, and preferably air is the original source of such mixtures. While the speed of reaction is somewhat reduced at low concentrations of either ethylene or oxygen, the mixtures otherwise give comparable chemical results.

It has been found that the velocity of the gases through the reaction zone does not exert a critical effect on the reaction, and the velocity need only be varied to give the degree of reaction desired, as correlated with the activity of the catalyst and the regulation of the temperature in the operation of the process.

4. CONCLUSION

From my knowledge of the Lefort process and experience with its operation, and from my knowledge of and experience with industrially used processes for the syntheses of aliphatic organic substances, I consider Lefort's invention to be one of the outstanding chemical discoveries in organic synthesis of recent years, and that it is both technically and economically suited for commercial operation and is of great industrial promise.

George O. Curme, Jr.

Sworn to and subscribed before me this 6th day of Apr 1937, at New York, N. Y.

E. Connell
Notary Public
NOTARY PUBLIC, New York County
New York County Clerk's No. 102
New York County Register's No. 8-D-139
Commission expires March 30, 1938

Filed Sept. 25, 1936

U. S. PATENT OFFICE
APR 10 1937
DIVISION

REPUBLIC OF FRANCE :
CITY OF PARIS : ss:
CONSULATE GENERAL OF THE UNITED STATES OF AMERICA :

L. GUICHARD, being duly sworn says:

I am a citizen of the Republic of France and reside in the City of Paris and I am a patent attorney associated with the firm of Elluin and Barnay. I maintain offices at 80 Rue Saint-Lazare, Paris, France. I have had no special academic or practical training in chemistry.

I acted as attorney for Société Francaise de Catalyse Generalisée in filing applications and obtaining various French patents on M. Lefort's process for obtaining ethylene oxide which is also the subject matter of United States Letters Patent No. 1,998,878, and I was solely responsible therefor.

After the various French patents had issued M. Lefort told me that he desired to protect his invention in the United States. I thereupon prepared, principally from French Patent No. 739,662, an application which I sent to Messrs. Marks & Clerk, National Press Building, 14th and F Streets, N.W., Washington, D. C., for filing in the United States Patent Office. This application, as prepared by me, was filed on March 22, 1932 and given Serial No. 600,578.

At the time that I drew the said application I had received no explanation of the nature of the invention other than in letters written to me by M. Lefort, one letter immediately preceding the filing of application for each of the various French patents. The content of each of these several letters appears substantially unchanged in the corresponding French patent. At the time I prepared the United States

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application I had no knowledge whatsoever of the state of the prior art.

From the first time when I concerned myself with patenting M. Lefort's process for the catalytic oxidation of ethylene to ethylene oxide and until after the issuance date of U. S. patent 1,998,878, I was under the mistaken belief that there was only one path by which ethylene could be oxidized, namely, through formaldehyde as an intermediate product. Apparently, M. Lefort, in turn, mistakenly believed that I knew his process accomplished direct reaction to ethylene oxide.

In June, 1932 I received a copy of German Patent No. 168,291, issued on March 15, 1906 to Dr. Johann Walter which was cited by the German Patent Office in an official action, dated June 22, 1932, on the German application for a patent on the Lefort process for the manufacture of ethylene oxide. This German patent to Walter describes the use of steam and metallic catalysts, silver among others, in the manufacture of formaldehyde by the oxidation of ethylene. Due to my mistaken understanding of the chemistry involved in the Lefort process I mistakenly assumed, after reading the said German patent, that the process, which was the subject matter of United States application Ser. No. 600,578, was like the process described in "Example 3", using the catalysts described at lines 105 to 113, page 3, of German Patent No. 168,291 except for Lefort's use of a special activated silver catalyst.

The United States Patent Examiner cited Willstatter, Annalen des Chemie, Vol. 422, pages 36 to 46 in the second official action. This is an article describing the manufacture of formaldehyde by the oxidation of ethylene. The citation of this article led me to believe that the Examiner also thought

that the formaldehyde process was the same as the Lefort process and I again mistakenly believed that this was the fact since I did not understand the real invention.

Solely by reason of my mistaken concept of the process of M. Lefort's invention and my mistaken and erroneous understanding of the true relation of his process to the prior art, I permitted the claims of Lefort's application Serial No. 600,578 to be restricted to narrow and specific embodiments of the invention therein described, and failed originally to draft claims of scope broader than the claims of said application as filed.

A. Guitzard.

Sworn to and subscribed before me this 30th day of March, 1937 at Paris, France.

[signature]

United States Consul

JOHN K. [illegible]
Vice-Consul of the United States
of America

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AMERICAN CONSULAR GENERAL
$2.00
FRS. 44.00
FRANCE

Filed Sept. 25, 1936 (3)

APR 10 1937

U. S. PATENT OFFICE
APR 12 1937
DIVISION 59

A F F I D A V I T

DISTRICT OF COLUMBIA)
) ss:
CITY OF WASHINGTON)

HERMAN JAKOBSSON, being duly sworn deposes and says:

THAT he resides at 228 Spruce Avenue, Takoma Park, D. C., and that he is an attorney duly registered and entitled to represent applicants before the Patent Office; that he was employed in the office of Marks & Clerk, National Press Building, 14th and F Streets, N.W., Washington, D. C. and in the office of the firm of Glascock, Downing & Seebold, of the same address, since the formation of the latter firm and until March, 1937; and that his successive employment by the two said firms covered the entire period during which the Lefort application, Serial No. 600,578 was pending in the Patent Office.

THAT he was solely responsible for the filing in the Patent Office of the said application Serial No. 600,578 and all amendments thereto; that he filed the said application exactly in the form in which it was received from Messrs. Elluin & Barnay, Paris, France; and that all amendments thereto were made and filed by deponent only upon authority or directions from Elluin & Barnay.

Deponent further says that he forwarded references cited in the prosecution of the said application to Elluin & Barnay for their consideration and that he did not attempt to understand the said invention and its relation to the cited

references with a view to distinguishing the said invention from the references cited; but that he left the consideration of the patentability of the said invention over the cited references wholly to Elluin & Barnay, and by arrangement with the latter acted only to transmit communications from and to the Patent Office.

Herman Jacobson

Sworn to and subscribed before me this 3rd day of April, 1937 at Washington, D. C.

[illegible]
Notary

APR 5 '37

IN THE UNITED STATES PATENT OFFICE

Div. 59, Room 4709 : Serial No. 102,613

T. E. Lefort : Filed Sept. 25, 1936

PROCESS FOR PRODUCING OLEFINE OXIDES

Hon. Commissioner of Patents

Washington, D. C.

Sir:

Supplementing the amendments to this application filed April 10, 1937, the following amendment is requested:

Claim 19, line 11, change "such as" to -- having the catalytic characteristics of --

Entire for allowance

REMARKS

The amendments to this application and the other papers filed April 10, 1937 were briefly discussed with the Examiner at a conference on that date, and again on April 15, 1937. The Examiner's courtesy in considering those papers and granting the further interview is acknowledged.

The foregoing amendment to claim 19 is made because of the Examiner's opinion expressed at the conference that the definition of the catalyst in the claim lacks adequate precision. This view is not believed to be completely valid, and claim 19 taken in the light of the specification is believed properly to define the catalytic materials for use in the process. However, the claim is being amended to a form which should remove this difference of opinion, and the amended form is submitted to be plainly definite in all respects. It is clear that an infringer cannot escape liability in the adoption of the process of claim 19 merely by replacing silver with an equivalent catalytic material otherwise within the class defined

79.

by the claim and pointed out in the description. There is no prior art cited, or known to applicant, which discloses applicant's process or any catalyst therefor. The invention is of pioneer character, and it is to be understood that applicant, by the present amendment, is not admitting any limitation on the scope of protection to which he is entitled. Applicant is endeavoring to comply with the formal requirements of the Examiner, and will look to the Courts later, if it becomes necessary, for interpreting the claims in the broad scope commensurate with the invention which has been made. Accordingly, applicant is willing to employ the amended language of this claim if it is more in conformity with the Examiner's viewpoint.

Other matters considered with the Examiner at the conference need not be restated here, since they are covered in full by the papers filed on April 10, 1937.

The original Letters Patent No. 1,998,878, surrender of which was tendered in the Petition for reissue of said patent submitted in this application, are enclosed. It is respectfully asked that this application be passed to issue at an early date.

Respectfully,

Eugene S. Greenwald
Attorney for Applicant

New York, N. Y.
April 15, 1937

F/D

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DEPARTMENT OF COMMERCE

UNITED STATES PATENT OFFICE

WASHINGTON

April 15, 1937.

Theodore Emile Lefort, Assor:

Sir:

Your application for Reissue for Patent No. 102,613 for improvement in process for the production of ethylene oxide, has been examined and allowed.

The patent will be forwarded to you as soon as practicable, in due order of business.

Very respectfully,

Thomas E. Robertson
Commissioner of Patents.

PATENT OFFICE
MAY 18 1937

Charles C. Scheffler
30 East 42nd Street,
New York, N. Y.

102613

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11—6041

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DEFENDANT'S EXHIBIT 6

FRENCH REPUBLIC

MINISTRY OF COMMERCE AND INDUSTRY

DEPARTMENT OF INDUSTRIAL PROPERTY

Patent of Invention

Group 14 — Class 1 No. 729,952

PROCESS FOR OBTAINING ETHYLENE GLYCOL

Societe Anonyme: SOCIETE FRANCAISE DE CATALYSE GENERALISEE, Residing in France (Seine)

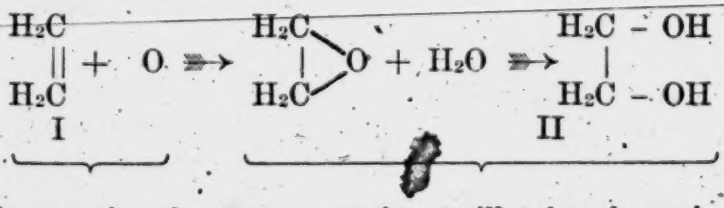
Filed March 27, 1931 at 4:20 p.m. in Paris

Granted May 3, 1932—Published Aug. 4, 1932.

(Patent of invention the grant of which has been deferred in accordance with article 11, paragraph 7 of the law of July 5, 1844 as amended by the law of April 7, 1902).

The invention has for its object a process for obtaining ethylene glycol which consists in subjecting ethylene derived from any source whatever; coke-oven gas, hydrogenation of acetylene, dehydration of ethyl alcohol, etc., to the simultaneous action of atmospheric oxygen and water.

The reaction underlying this process may be envisaged in the following manner:



In practice these two reactions will take place simultaneously at a temperature of 150 to 250° C. and a minimum pressure of 50 kg. although tests have shown that the reaction took place at atmospheric pressure.

As a catalyst one may use either antimony, or arsenic, or their compounds, or a mixture of these two elements and of their compounds.

Method of operation:—One may use high pressure steel equipment having two compressors, one for compressing air and the other for compressing ethylene, although it does not a priori appear impossible to compress a mixture of these two gases in one and the same compressor by keeping the mixture outside the explosive limits.

These two gases are passed into a reaction tube containing one of the aforementioned catalysts and heated to a temperature of about 200° C., the reaction tube being such that it will withstand a pressure of several hundred kilograms. This tube is moreover connected with a circulation pump permitting the introduction of water under pressure.

The reaction between the ethylene, atmospheric oxygen and water will take place in this tube. A continuous operation may be achieved by regulating the rates of flow. The glycol thus formed may be separated from the water by simple distillation.

Resumé

1. Process for obtaining ethylene glycol by the simultaneous action of atmospheric oxygen and water on ethylene;

2. This reaction is preferably carried out under a minimum pressure of 50 kilograms; the pressure may reach several hundred kilograms without inconvenience;

3. The reaction temperature is preferably maintained between 150 and 300° C.;

4. The catalysts used are particularly arsenic, antimony, their compounds, their mixtures, or mixtures of the latter;

5. By avoiding the addition of water to the reaction one can obtain solely ethylene oxide.

Societe Anonyme: SOCIETE FRANCAISE DE CATALYSE GENERALISEE

By power of attorney:
Elluin and Barnay.

FRENCH REPUBLIC

MINISTRY OF COMMERCE AND INDUSTRY

DEPARTMENT OF INDUSTRIAL PROPERTY

First Addition
To Patent of Invention
No. 729,952

Group 14 — Class 1 No. 41,255

PROCESS FOR OBTAINING ETHYLENE GLYCOL

Societe Anonyme: SOCIETE FRANCAISE DE CATALYSE GENERALISEE residing in France (Seine)

(Principal patent applied for March 27, 1931)

Filed July 4, 1931 at 2:12 p.m. in Paris.

Granted August 30, 1932—Published Dec. 2, 1932.

(Certificate of addition the grant of which has been deferred in accordance with article 11, paragraph 7 of the law of July 5, 1844 as amended by the law of April 7, 1902).

The object of the process of the principal French patent for "Process of obtaining ethylene glycol", dated March 27, 1931, consists in subjecting ethylene to the simultaneous actions of oxygen and water to obtain ethylene glycol.

In one method of carrying out this process air, ethylene and water are introduced under pressure into a chamber containing a catalyst and heated to about 200° C.

The catalysts referred to in the principal patent are antimony, arsenic, or their compounds, alone or in admixture.

The present addition has for its object the disclosure of other catalysts which in the course of experimentation have been found to be efficacious.

These various catalysts are the following:

1. A mixture of antimony, bismuth, silver, lead, arsenic, nickel and tin in variable proportions.

2. Bismuth alone.
3. Mixtures of antimony, lead, nickel and silver.
4. Mixtures of bismuth, nickel, silver and lead.
5. Mixtures of antimony, bismuth, nickel.
6. Mixtures of antimony, bismuth, silver.
7. Mixtures of bismuth and nickel.
8. Mixtures of bismuth and silver.

And in general any mixture that can be made from the following metals: Antimony, lead, bismuth, silver, nickel, tin, arsenic, taken alone, or taken 2 by 2, 3 by 3, 4 by 4, etc., in variable proportions; mixtures, under the same conditions, of these metals with their oxides; mixtures of only the oxides of these metals under the same conditions.

Resumé

The object of the present addition is to indicate new catalysts suitable for carrying out the "Process for obtaining ethylene glycol" which forms the object of the principal patent dated March 27, 1931.

These catalysts are in general:

Any mixture that can be made from the following metals: antimony, lead, bismuth, silver, nickel, tin, arsenic, taken alone, taken 2 by 2, 3 by 3, 4 by 4, etc., in variable proportions.

Mixtures of these metals and their oxides under the same conditions.

Mixtures of only the oxides of these metals under the same conditions.

Societe anonyme: SOCIETE FRANCAISE DE CATALYSE GENERALISEE

By Power of attorney:
Elluin and Barnay.

FRENCH REPUBLIC

MINISTRY OF COMMERCE AND INDUSTRY

DEPARTMENT OF INDUSTRIAL PROPERTY

Second Addition
To Patent of Invention
No. 729,952

Group 14 — Class 1 No. 41,474

PROCESS FOR OBTAINING ETHYLENE GLYCOL

Societe Anonyme: SOCIÉTE FRANCAISE DE CATALYSE GENERALISEE residing in France (Seine)

(Principal patent applied for March 27, 1931).

Filed September 10, 1931 at 4:13 p.m. in Paris

Granted Oct. 25, 1932—Published January 10, 1933.

(Certificate of addition the grant of which has been deferred in accordance with article 11, paragraph 7 of the law of July 5, 1844 as amended by the law of April 7, 1902).

First addition No. 41,255.

The principal French patent dated March 27, 1931, for "Process for obtaining ethylene glycol" consists in subjecting ethylene to the simultaneous action of oxygen and water to obtain ethylene glycol.

The object of the first addition, dated July 4, 1931, to the said principal patent is to indicate catalysts other than those mentioned in the principal patent.

The purpose of the present addition is to specify that the formation of glycol obtained by the oxidation of ethylene by atmospheric oxygen, in the presence of water, is necessarily derived from the saponification of the ethylene oxide obtained in the first stage by direct oxidation of the ethylene. The mechanism, indicated in the principal patent and the first addition relative thereto, accordingly leads first to the production of ethylene oxide which by direct saponification gives glycol.

But tests made by the applicant show that the catalysts indicated do not enable one to obtain ethylene oxide by the oxidation of ethylene if the gases are dry. The presence of water is necessary for the production of ethylene oxide, even apart from any attempt to bring about saponification.

The mechanism for obtaining glycol from ethylene as indicated above is specified below by way of example. Silver in the form of a powder or in the form of gauze, either on a support or not, is placed in a tube, for example of silica, and after having heated the tube to a temperature between 250 and 400°, a mixture of ethylene, air and water vapor is passed therethrough. The gases coming out of the tube are passed into a series of wash bottles in order to absorb the ethylene oxide formed by the reaction. The liquid in the wash bottles is then saponified, either with or without catalysts, to obtain glycol.

A silver colloid can also be introduced into a high pressure tube which has been filled with water. Pure ethylene is then added under pressure in order to dissolve it in the water.

The tube is then heated to between 150 and 300° C. whereupon air is slowly introduced. When all the ethylene is transformed into ethylene oxide, a saponification is effected so as to obtain the glycol.

Resumé

The present addition to the principal French patent dated March 27, 1931, for "Process for obtaining ethylene glycol" and the first addition dated July 4, 1931, is characterized principally by the following points:

1. Obtaining glycol by the production of ethylene oxide and saponification.

2. Obtaining ethylene oxide by direct oxidation of ethylene by atmospheric oxygen in the presence of water vapor and in contact with the catalysts specified in the principal patent and in the first addition, particularly silver and its mixtures with bismuth, antimony, lead, tin, nickel.

3. The necessity of introducing water vapor into the reacting gases (ethylene and air) to obtain ethylene oxide in accordance with the processes indicated above.

4. Obtaining ethylene oxide and then glycol in the liquid phase in a pressure tube in the presence of colloidal silver, taking advantage of the high solubility of ethylene in water under pressure.

Societe Anonyme: SOCIETE FRANCAISE DE CATALYSE GENERALISEE

By Power of Attorney:
Ellain and Barnay.

FRENCH REPUBLIC

MINISTRY OF COMMERCE AND INDUSTRY

DEPARTMENT OF INDUSTRIAL PROPERTY

Third Addition

TO PATENT OF INVENTION

No. 729,952

Group 14 — Class 1. No. 41,484

PROCESS FOR OBTAINING ETHYLENE GLYCOL

Societe Anonyme: SOCIETE FRANCAISE DE CATALYSE GENERALISEE Residing in France (Seine)

(Principal patent applied for March 27, 1931)

Filed September 25, 1931 at 10:45 a.m. in Paris
Granted November 3, 1932—Published January 10, 1933
(Certificate of addition the grant of which has been deferred in accordance with article 11, paragraph 7 of the law of July 5, 1844 as amended by the law of April 7, 1902).

First Addition No. 41,255 Second Addition No. 41,474

The process which is the object of the principal French patent for "Process for obtaining ethylene glycol" dated March 27, 1931, consists in subjecting ethylene to the simultaneous action of oxygen and water in the presence of a catalyst to obtain ethylene glycol.

Various catalysts suitable for carrying this process into practice have been disclosed in the above principal patent and in its first addition.

A method of operation described in a second addition consists in passing a mixture of ethylene, air and water vapor into a tube of refractory material containing the catalyst and heated to a temperature of between 250 and 400°. On leaving the tube, the gaseous mixture is passed into wash bottles the liquid of which is then saponified to obtain glycol.

The present addition has for its object an improvement in the process of the principal patent. This improvement consists in introducing hydrogen into the gaseous mixture of ethylene, air and water vapor.

The quantity of hydrogen introduced is at least 5% of the total mixture.

The reaction is thus greatly facilitated and the yield considerably increased. The proportion of hydrogen may be varied with experimental conditions, and particularly with the temperature and pressure.

Resumé

The present addition to the principal patent for "Process for obtaining ethylene glycol", dated March 27, 1931, has for its object an improvement in this process characterized in that hydrogen is introduced into the gaseous mixture of ethylene, air and water vapor, the passage of which into a heated chamber in the presence of a catalyst brings about the basic reaction of the process.

The proportion of hydrogen may be varied with the temperature and pressure. It constitutes at least 5% of the total gaseous mixture.

Societe Anonyme: SOCIETE FRANCAISE DE CATALYSE GENERALISEE

By Power of attorney:
Elluin and Barnay.

FRENCH REPUBLIC
MINISTRY OF COMMERCE AND INDUSTRY
DEPARTMENT OF INDUSTRIAL PROPERTY

Fourth Addition
To Patent of Invention
No. 729,952

Group 14 — Class 1. No. 41,810

PROCESS FOR OBTAINING ETHYLENE GLYCOL

Societe anonyme dite: SOCIETE FRANCAISE DE CATALYSE GENERALISEE residing in France (Seine)

(Principal patent applied for March 27, 1931)

Filed December 19, 1931 at 4:47 p.m. in Paris
Granted January 21, 1933—Published April 21, 1933

(Certificate of addition the grant of which has been deferred in accordance with article 11, paragraph 7 of the law of July 5, 1844 as amended by the law of April 7, 1902).

First Addition No. 41,255
Second " No. 41,474
Third " No. 41,484

The principal French patent dated March 27, 1931, for "Process for obtaining ethylene glycol", has for its object a process for obtaining ethylene glycol which consists principally in subjecting ethylene to the simultaneous action of atmospheric oxygen and water in the presence of catalysts under atmospheric or superatmospheric pressure.

Among the catalysts referred to is silver, and in the third addition to the principal patent there is reference to the introduction of hydrogen into the mixture of ethylene, air and water vapor in order to facilitate the reaction.

The present addition has for its object modifications and improvements in the process described in the prin-

cipal patent characterized by the following points, applicable either separately or in combination:

a. The silver is activated by introducing thereinto small quantities of gold, copper and iron;

b. As catalysts one may use the metals: gold, copper, iron, mixed or not, and in all proportions;

c. The yield of the reaction is increased by diminishing the CO_2 which is formed, by introducing a suitable quantity of water into the reaction, and by initially admixing a suitable volume of CO_2 with the gaseous reactants;

d. To improve the yield of ethylene oxide the synthesis is carried out under a pressure less than atmospheric.

Experiments have in fact shown that silver can be greatly activated by the introduction of small quantities of gold, copper and iron.

Thus, a mixture containing silver, gold, copper and iron gives very good results and permits of a specific reaction; the only foreign product obtained in the reaction is CO_2 which is easily removed. Only traces of aldehydes are formed.

The catalytic mixtures may comprise

Silver, gold, copper, iron Silver, gold, copper Silver, gold Silver, copper, iron Silver, copper Silver, iron Copper, iron	In all Proportions.

As indicated above, one may use as catalysts the metals: gold, copper, iron, either in admixture or not and in all proportions.

Moreover, one has established that the reaction producing the CO_2 is, contrary to one's previous belief, an oxidation reaction independent of that which produces ethylene oxide and of that which produces aldehydes.

From experiments of the applicant, it results that if water in suitable quantity is introduced, not only is the reaction facilitated, as indicated in the third addition, but in addition, the reaction producing CO_2—probably by direct oxidation of ethylene in accordance with the following equation:

$$C_2H_4 + 3O_2 = 2H_2O + 2\,CO_2$$

is checked, due—probably—to the partial pressure of the water. From these experiments one may conclude that if a certain quantity of CO_2 is introduced, in addition, initially into the reacting gases, the partial pressure of the CO_2 will intervene and the reaction by which ethylene is oxidized to CO_2 will be completely checked, resulting in a considerable increase in the yield of the operation. Moreover, it is established that when the synthesis is carried out under reduced pressure the yield of ethylene oxide is improved.

Resumé

The present addition to the principal patent dated March 27, 1931, has for its object certain modifications and improvements in the process described in the principal patent and in the third addition thereto, characterized especially by the following points, applicable separately or in combination:

a. Silver is activated by introducing thereinto small quantities of gold, copper and iron;

b. As catalysts one may use the metals: gold, copper, iron, either mixed or not and in all proportions;

c. The yield of the reaction is increased by diminishing the CO_2 which is formed, by introducing a suitable quantity of water into the reaction, and by initially admixing a suitable volume of CO_2 with the reacting gases.

d. To improve the yield of ethylene oxide the synthesis is carried out under sub-atmospheric pressure.

Societe Anonyme dite: SOCIETE FRANCAISE DE CATALYSE GENERALISEE

By Power of attorney:
Elluin and Barnay.

FRENCH REPUBLIC
MINISTRY OF COMMERCE AND INDUSTRY
DEPARTMENT OF INDUSTRIAL PROPERTY

Patent of Invention
No. 739,562

Group 14 Class 1

PROCESS FOR OBTAINING ETHYLENE OXIDE, PARTICULARLY FOR THE PURPOSE OF PREPARING ETHLYENE GLYCOL AND ITS DERIVATIVES

Societe Anonyme dite: SOCIETE FRANCAISE DE CATALYSE GENERALISEE residing in France

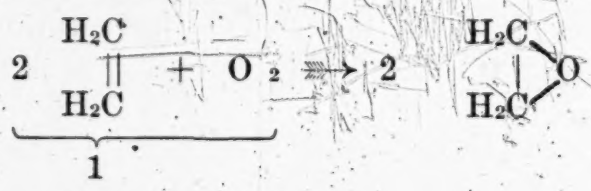
(Seine)

Filed October 3, 1931 at 4:30 p.m. in Paris
Granted November 3, 1932—Published January 13, 1933

(Patent of invention the grant of which has been deferred in accordance with article 11, paragraph 7 of the law of July 5, 1844 as amended by the law of April 7, 1902).

The present invention has for its object a process for obtaining ethylene oxide which consists principally in subjecting ethylene to the simultaneous action of atmospheric oxygen and water in the presence of a catalyst and hydrogen.

The hydrogen probably acts as an accelerating agent for the basic reactions of the process which may be expressed in the following form:



This reaction takes place at atmospheric pressure but for the industrial application of the process the pressure may be, for example, 50 kilograms per square centimeter at a temperature of between 150 and 400° C.

The following catalysts may be employed:

1. A mixture of antimony, bismuth, silver, lead, arsenic, nickel and tin in variable proportions.

2. Silver alone;
bismuth alone;
antimony alone.

3. Mixtures of antimony, lead, nickel and silver.

4. Mixtures of bismuth, nickel, silver and lead.

5. Mixtures of antimony, bismuth, nickel.

6. Mixtures of antimony, bismuth, silver.

7. Mixtures of bismuth and nickel.

8. Mixtures of bismuth and silver.

And, in general, any mixture that can be made from the following metals: antimony, lead, bismuth, silver, nickel, tin, arsenic, taken alone, taken two by two, three by three, four by four, etc., in variable proportions; mixtures of these metals with their oxides, under the same conditions; mixtures of only the oxides of the metals, under the same conditions.

The ethylene can be derived from any source; coke oven gas, hydrogenation of acetylene, dehydration of ethyl alcohol; and others.

Water can be introduced into the reaction chamber either in the liquid state or as steam.

The oxygen can be the oxygen of the air, this latter gas being introduced in the reaction.

Described below, by way of example only, are various methods of carrying out the process:

Method of operation: 1.—In an apparatus capable of resisting high pressures one compresses separately by means by suitable compressors, ethylene and air.

A mixture of air and ethylene may also be compressed, all necessary precautions being taken to prevent an explosion.

The two gases, together with hydrogen, are passed into a reaction tube resistant to pressures of several

hundred kilograms containing one of the catalysts indicated above and heated to a temperature, for example, of 200° C. The tube is also connected with a circulation pump permitting the introduction of water under pressure.

The reaction between the ethylene, the oxygen of the air, and the water takes place in the tube. Continuous operation may be had by regulating the rates of flow. The ethylene oxide thus formed is separated from the other gases, for example, by condensation.

Method of operation: 2.—Silver in the form of a powder, or in the form of gauze, either supported or not, is introduced into a tube made, for example, of silica, and after heating the tube to a temperature between 150 and 400° C., a mixture of ethylene, air, water vapor and hydrogen is passed therethrough. The proportion of the latter gas amounts to at least 5% of the total mixture. The ethylene oxide produced may be separated from the gases which emerge from the tube.

Method of operation: 3.—Colloidal silver is introduced into a high pressure tube filled with water. Pure ethylene is then added under pressure in order to dissolve it in the water.

The tube is then heated to between 150 and 400° C., whereupon air and hydrogen are slowly introduced. The introduction of air is stopped when all the ethylene has been transformed into ethylene oxide.

From the ethylene oxide produced in accordance with the above processes, one may obtain ethylene glycol by simple saponification, or by other known methods any derivatives of ethylene glycol.

The reaction for producing ethylene glycol may be expressed as follows:

$$C_2H_4O + H_2O \Longrightarrow \begin{matrix} H_2C - OH \\ | \\ H_2C - OH \end{matrix}$$

Tests made by the applicant have shown that in the presence of the catalysts specified, water, in the form of

steam or not, considerably favors the reaction of the production of ethylene oxide.

Resumé

The invention has for its object a process for producing ethylene oxide characterized principally in that ethylene is subjected to the simultaneous action of atmospheric oxygen and water in the presence of a catalyst and hydrogen.

The catalysts suitable for carrying out the process are all the mixtures that can be made from the following metals: antimony, lead, bismuth, silver, nickel, tin, arsenic, taken alone, taken two by two, three by three, four by four, etc., in variable proportions.

Mixtures of these metals with their oxides under the same conditions.

Mixtures of only the oxides of these metals under the same conditions. Only the oxides of the above mentioned metals.

The reaction takes place at atmospheric pressure but it may be carried out advantageously under pressure at a temperature between 150 and 400° C.

One method of operation consists in passing a mixture of ethylene, air, water vapor and hydrogen into a tube of refractory material containing the catalyst, the tube being heated to a temperature between 150 and 400° C. On leaving the tube the gaseous mixture is passed into condensers which permit collecting the ethylene oxide produced.

The quantity of hydrogen introduced amounts to at least 5% of the total mixture.

From the ethylene oxide one may obtain ethylene glycol by simple saponification, or by any suitable treatment one may obtain the derivatives of ethylene glycol.

Societe Anonyme dite: SOCIETE FRANCAISE DE CATALYSE GENERALISEE

By Power of attorney
Elluin and Barnay.

FRENCH REPUBLIC
MINISTRY OF COMMERCE AND INDUSTRY
DEPARTMENT OF INDUSTRIAL PROPERTY

First Addition
To Patent of Invention
No. 739,562

Group 14 — Class 1 No. 41,811

PROCESS FOR OBTAINING ETHYLENE OXIDE, PARTICULARLY FOR THE PURPOSE OF PREPARING ETHYLENE GLYCOL AND ITS DERIVATIVES

Societe anonyme dite: SOCIETE FRANCAISE DE CATALYSE GENERALISEE residing in France (Seine)

(Principal patent applied for October 3, 1931)

Filed December 19, 1931 at 4:48 p.m. in Paris
Granted January 21, 1933—Published April 21, 1933.

(Certificate of addition the grant of which has been deferred in accordance with article 11, paragraph 7 of the law of July 5, 1844, as amended by the law of April 7, 1902).

The process described in the principal French patent for a "Process for obtaining ethylene oxide particularly for the purpose of preparing ethylene glycol and its derivatives", dated October 3, 1931, consists principally in subjecting ethylene to the simultaneous action of oxygen and water in the presence of a catalyst and hydrogen.

In the examples for carrying out this process described in the principal patent, the reaction takes place at atmospheric or superatmospheric pressure in the presence of catalysts among which is mentioned silver in colloidal form, or in the form of powder or gauze.

The present addition has for its object modifications and improvements in the process of the principal patent characterized by the following points, applicable separately or in combination:

a. The silver is activated by introducing thereinto small quantities of gold, copper, and iron;

b. As catalysts one uses the metals: gold, copper, iron, either in admixture or not, and in all proportions;

c. The yield of the reaction is increased by diminishing the CO_2 which is formed, by introducing a suitable quantity of water into the reaction and by initially admixing with the reacting gases a suitable volume of CO_2;

d. The synthesis is carried out under less than atmospheric pressure to improve the yield of ethylene oxide.

Experiments have, in fact, shown that silver may be activated by the introduction into this substance of small quantities of gold, copper and iron.

Thus a mixture containing silver, gold, copper and iron gives very good results and permits of a specific reaction; the only foreign product obtained in the reaction is CO_2 which may be easily removed. Only traces of aldehydes are formed.

The catalytic mixture may comprise

Silver, gold, copper, iron Silver, gold, copper Silver, gold Silver, copper, iron Silver, copper Silver, iron Copper, iron	In all proportions

As indicated above, one may use as catalysts the metals: gold, copper, iron, either in the form of mixtures or not, and in all proportions.

Moreover, one has established that the reaction producing the CO_2 is, contrary to one's previous belief, an oxidation reaction independent of that which produces ethylene oxide and of that which produces aldehydes. From experiments of the applicant, it results that if water

in suitable quantity is introduced, not only is the reaction facilitated, as indicated in the principal patent, but in addition, the reaction producing CO_2—probably by direct oxidation of ethylene in accordance with the following equation:

$$C_2H_4 + 3O_2 = 2H_2O + 2CO_2$$

is checked, due—probably—to the partial pressure of the water. From these experiments one may conclude that if a certain quantity of CO_2 is introduced, in addition, initially into the reacting gases, the partial pressure of the CO_2 will intervene and the reaction by which ethylene is oxidized to CO_2 will be completely checked, resulting in a considerable increase in the yield of the operation. Moreover, it is established that when the synthesis is carried out under reduced pressure the yield of ethylene oxide is improved.

Resumé

The present addition to the principal patent dated October 3, 1931, has for its object certain modifications and improvements in the process described in the principal patent, characterized especially by the following points, applicable separately or in combination:

a. Silver is activated by introducing thereinto small quantities of gold, copper and iron;

b. As catalysts one may use the metals: gold, copper, iron, either mixed or not and in all proportions;

c. The yield of the reaction is increased by diminishing the CO_2 which is formed, by introducing a suitable quantity of water into this reaction, and by initially admixing a suitable volume of CO_2 with the reacting gases;

d. To improve the yield of ethylene oxide the synthesis is carried out under subatmospheric pressure.

Societe anonyme dite: SOCIETE FRANCAISE DE CATALYSE GENERALISEE,

By Power of attorney:
ELLUIN AND BARNAY.

DEFENDANT'S EXHIBIT 42

Eighteenth
May
1933

E. L. Masthoff, Esq.,
De Bataafsche Petroleum Mij.,
30 Carel van Bylandtlaan,
The Hague, Holland.

My dear Masthoff:

The Shell Development laboratory at Emeryville is in correspondence with the firm: Givaudan-Delawanna Inc., 80 Fifth Avenue, New York, who may be interested in products like Methyl-Allyl-Chloride, Iso-Crotyl-Chloride, 2-3 Dichlor-Iso-Butane, and normal Propyl-Chloride, they also report that they are interested in 40,000 to 70,000 lbs. of Chlorhydrene. Mr. Steck, who is going East shortly will discuss these subjects with them.

In one of their letters they write the following:

> "Since I wrote you my first letter, I have received news from abroad that your good company has taken over the patent rights for the manufacture of *Ethylene Oxide* according to the process patented by the French Company "Societe de Catalyse Generalisee". This process, I understand, allows you to make Ethylene Oxide directly from Ethylene without going through the Chlorhydrene and it might well be that under such circumstances it might be advisable for us to use the Ethylene oxide provided it can be obtained at a reasonable price. On the other hand, you may not want to make it unless in very large quantities, and should this be the case, I am wondering whether you would be willing to let us have a license, that is, a manufacturing license only."

Will you please let us know:

Firstly: Did B.P.M. actually acquire this French patent No. 729,952?

Secondly: If so, have U.S. applications been made corresponding to this French patent?

Thirdly: And if so, has B.P.M. acquired also the rights to the U.S. patent applications?

Fourthly: Have you any information regarding actual results obtained by the process covered by the French patent 729,952?

If the process is good, it should be very attractive and we should like to work on its development. We have plenty of ethylene available at Shell Point for carrying out the necessary experiments on a semi-commercial scale.

Yours sincerely,

DP:B

DEFENDANT'S EXHIBIT 43

E.L.MASTHOFF

'S-GRAVENHAGE, 28th June 1933
CAREL VAN BYLANDTLAAN 30

AIRMAIL. 555

D. PIJZEL, Esq.,
c/o Shell Development Company,
100 Bush Street,
SAN FRANCISCO.

Cal., U.S.A.

CI (CC-OA) — June 28, 1933

My dear Pijzel,

My reply to your letter D N° 215 of the 18th May last has been delayed as we were just having conferences with the gentlemen of the "Société de Catalyse Généralisée" regarding their process of making ethylene oxide.

First of all, I shall answer your questions:

Firstly: B.P.M. have not acquired rights on the French patent N° 729.952, nor on any of the 4 subsequent French patents, nor on any other foreign application.

Secondly: One U.S. application comprising all the French patents has been filed, of which you will find a copy, translated into French, enclosed. We have been promised the original application, which will be sent to you on receipt.

Thirdly: see under firstly.

Fourthly: We have information on the process and its results in laboratory tests only. This information is contained in the enclosed copy of Dr Langedijk's report N° 5205.

We obtained the first details regarding this French process from the Hercules Powder Co's representative in Holland, Mr Riemersma. He told us that the Hercules Co in U.S.A. are interested in ethylene glycol for the manufacture of explosives but that the price Hercules buy it for, viz. ¢ 15/lb was too high for a more general application. As the price quoted in the market reports of American chemical periodicals is ¢ 25/lb, it seems to me that Hercules are either buying at a large discount or that Mr Riemersma kept his price information too low for some reason or other. Riemersma told us that the French patent N° 729.952 had attracted his attention and he had arranged a visit to the laboratoy of the Soc. de C. G., where he had gained a favourable impression of the process demonstrated to him on a laboratory scale, especially as the French people had informed him that the cost price, inclusive of capital charges, by their process was ¢ 5/lb. glycol.

As Riemersma was shortly going to the U.S.A., he intended to discuss with his Directors the process and a prospective acquirement of the U.S.A. rights by the Hercules Co. However, as Hercules had no ethylene and the Shell had and seeing that his company and ours were already working together in some respects, he intimated that perhaps the Shell would care to obtain these

rights and manufacture the glycol under some agreement for Hercules, or Hercules would buy the rights and have glycol made under these rights by Shell, or Shell and Hercules would buy the rights together. Anyway he asked us to study the process and arranged a visit of one of our Amsterdam chemists to the Soc. de C. G. The outcome of this visit is laid down in Dr. Langedijk's report in which he describes the process followed on a laboratory scale.

You will see from this report that the ethylene—in this case ethylene 97%—is oxidized in mixture with the tenfold quantity of air and certain quantities of steam at ordinary pressure and a temperature of abt 375°C over a catalyst. To all appearances the catalyst used is silver activated with small quantities of gold, copper and iron. The reaction gas from each pass, after extraction of C_2H_4O and part of the CO_2 by waterwash is, without any subsequent admixture, recycled through the apparatus. In three subsequent passes of the run witnessed by Langedijk, of 36.3 L ethylene originally contained in the fresh feed, 31.82 L C_2H_4 had reacted (3.8 L C_2H_4 being retained in the gas holder for reaction gas, and 0.68 L C_2H_4 still contained in the final end gas). According to the description in the report of the 3 stages of run 1 and taking the method of determination of the oxide as glycol to be the more trustworthy one, it appears that these 31.82 L are divided over the reaction products as follows:

11.31 L C_2H_4 as oxide	=	35.5%
11.95 " " " CO_2	=	37.5%
8.56 " " not accounted for	=	27.–%
		100.–%

As you will see from the report, Dr. Langedijk who himself analysed on products of side reactions in the ethylene oxidation, did not find any other compounds besides C_2H_4O and CO_2. It seems therefore that the late Prof. Henry, who was the inventor, succeeded in developing a catalyst which drives the reaction between C_2H_4 and oxygen into one specified direction, whereby the ethylene in about equal parts in converted to the oxide and CO_2 respectively.

As no side reactions can be detected, the Sᵉ de C. G. feel qualified to calculate their % oxide yields as set forth on p. 9 of the report, viz:

$$R = \frac{\text{ethylene converted to oxide}}{\text{ethylene to oxide} + \text{ethylene to } CO_2} \times 100$$

thereby neglecting the amount of C_2H_4 not accounted for, as they say this is due to mechanical losses in their laboratory equipment and inaccuracy of analysis methods.

Although Dr Langedijk agrees that by evading such losses by better experimentation, the % of ethylene not accounted for, should be materially decreased, it seems in my opinion that such a high loss of about 27% cannot easily be ascribed to leakages in such a rather simple apparatus under very little more than ordinary pressure, and I think it highly important to have this point cleared up.

If it should be confirmed that only mechanical losses are responsible for the deficiency in ethylene, and therefore the process really converts the C_2H_4 to oxide and CO_2 only, so that the above Soc. de C. G.'s yield formula would be correct, a yield of about 45% of oxide introduced (see p. 9 of the report) may be reckoned upon.

This is important according to Mr Franchon, as the I.G. who are negotiating for the acquirement of the process rights for the European countries, should have stated that the French process could only compete with the Goldschmidt chlorhydrine process if yields of at least 40% on ethylene introduced would be obtained.

As you will see from the enclosed minutes we had a conference with two gentlemen of the Soc. de C. G. at The Hague, following a preliminary discussion between them, Mr C. B. de Bruyn and myself in Paris.

The gist of this conference was that BPM. have obtained a six months' option on the rights applied for in the U.S.A. The first thing for Shell Development Co. to do would be to study the process from information furnished to them by B.P.M., thereafter advising B.P.M.

as soon as possible whether Shell Development after this study would still be interested in the process. If so, results of the experiments with Shell Dev. Gases, which in order not to lose time should immediately be sent to Amsterdam, would then be made.

As regards costprice for the oxide made in this French process, you will see that we have only the assertion by Mr Franchon that the maximum costprice for French conditions would be frs. 3.— per kg. glycol, based on a price of frs 0.66 per m3 ethylene (calculated at 100%).

As stated in the minutes sulphur compounds are detrimental to the catalyst's activity, but Mr Franchon asserts that he is able to purify the gases sufficiently to prevent such poisoning. If it should be decided, therefore, to send other gases containing C_2H_4 besides your Shellpoint ethylene fraction, it is not necessary to have them purified on your side.

For the experiments in Paris and presumably in Amsterdam 3 cylinders of 40 kg each for each kind of gas to be examined would be required. These gases should be sent as soon as possible and we should appreciate it to receive an analysis of the gases shipped. Please mark the cylinders CI . 58

It is rather strange that Dr Kunz of the Givaudan Delawanna Corp. should have submitted the information to you that Shell Development Co. had taken over the patent rights on the Soc.'s process, which creates the impression that possibly the Soc. are trying to play off our Company against third parties interested in the process for U.S., especially as the French gentlemen intimated here that Dupont de Nemours were interested too.

I may mention here that we have not heard from Mr Riemersma since his recent return from the U.S.A.; however, we shall have a talk with him within a few days and shall keep you posted about what he has to say regarding his Company's point of view on glycol production according to the process of the Soc. de C. G.

I also enclose a note from our OA Department on the patent side of the process for the guidance of your patent department. We should be pleased if you would cable

us your ideas together with the date of shipment of the cylinders of gases, in case you wish to have experiments made.

Awaiting your reply with much interest, I remain,

yours sincerely,

E. L. MASTHOFF.

Encls: (*in duplicate*)

translation U.S.A. application. (one copy)
Report N° 5205 by Dr Langedijk.
Minutes of discussions.
Memo from our OA dept.
The 5 French patents

cc. Mr De Bruyn.

P. S. The memo from our O.A. Dept. has not been finished yet, and will be sent you by next mail. I enclose 2 copies of the original U.S.A. application 600,578, just received.

E. L. M.

DEFENDANT'S EXHIBIT 46

TRANSLATION.

B.P.M. LABORATORY REPORT NO 5205.

TITLE: Report on a visit to the "Société francaise de Catalyse généralisée" at Paris-Nanterre on March 30th and 31st and April 8th 1933 to judge their process for manufacturing ethylene oxide and glycol.

AUTHOR: Dr S.L. Langedijk, B.P.M. laboratory, Amsterdam.

SUMMARY: A description is given of the experiments made by the S.F.C.G. for converting ethylene-air mixtures in the

presence of a catalyst discovered by the S.F.C.G. at a temperature of 375°C. and at practically atmospheric pressure.

CONCLUSION: Starting from a mixture of practically pure ethylene and air in the ratio 1 : 10, this can be converted in three cycles through the apparatus to such an extent that there is practically no ethylene left. In the first cycle 2,5 gr. steam is added per liter gas mixture, in the second 1,4 gr. and in the third 0,7 gr. No formaldehyde, acetaldehyde, formic acid or carbon monoxide are formed during the reaction. Exclusively ethylene oxide (abt 40%), CO_2 (abt 40%) and a trace of glycol are found as reaction products. With more accurate methods of analysing and more expert experimenting the part not accounted for may certainly be reduced to much less than the 20—30% found in this case.

The S.F.C.G. method gives a better yield in oxide than the direct oxidising methods described so far; it remains to be proved by further experiments and careful calculations whether the S.F.C.G. method is preferable to the chlorhydrine method and whether it is directly suitable for technical ethylene.

Anticipating a consideration of the process from a patent technical point of view by the Patent Department, the author states as his personal opinion that the only characteristic feature of the process is the catalyst found by the S.F.C.G.; the activity and selectivity of the catalyst determine the other reaction condition, each of which (though having less effect) has already been mentioned in publications for the oxidation of ethylene with air or oxygen.

REPORT NO. 5205.

REPORT ON A VISIT TO THE "SOCIÉTÉ FRANÇAISE DE CATALYSE GÉNÉRALISÉE" AT PARIS-NANTERRE ON MARCH 30TH AND 31ST AND APRIL 8TH 1933 TO JUDGE THEIR PROCESS FOR MANUFACTURING ETHYLENE OXIDE AND GLYCOL.

The process for manufacturing ethylene oxide by oxidation of ethylene with air described in this report was demonstrated to me by the "Société Francaise de Catalyse Généralisée (abbreviated S.F.C.G.) in their laboratory at Nanterre, about 15 Km. west of Paris.

The Société Lyonnaise d'Electricité et d'Eau" (abbr. S.L.E.E.) have large gasworks with coke plant at Nanterre; the S.F.C.G. had rented the laboratory from the S.L.E.E., but otherwise these two companies seemed to be only very loosely associated.

In their patents concerning the manufacture of ethylene oxide and glycol by direct oxidation of ethylene (contingently in the presence of water) with air[1], the S.F.C.G. describe many methods of performing their process. However, when demonstrating the process to me, they omitted much of what they claim in their patents to be of advantage: instead of applying high pressure they worked at practically atmospheric pressure; the reaction took place at a temperature considerably over 300°C.; no hydrogen was added to the gas mixture; etc. In reply to a direct question whether this manner of working did not give an unsatisfactory idea as to the yield attainable, I was informed that the method followed for the demonstration yielded practically as much oxide as the modifications described in the patents. All that was absolutely necessary was:

a. to add an ample quantity of steam during the reaction (which, contrary to what is stated in the patents, gives rise to the formation of only very little glycol);

[1] So far only the French patents Nos. 729,952 and 739,562 of the S.F.C.G. concerning this subject have been published.

b. to use the specific catalyst discovered at the time by the late Prof. Ch. Henry, in regard to which no information at all was to be obtained.

It was further desirable:

c. to use a mixture of ethylene and air in the ratio 1 : 10 i.e. ethylene : $O_2 = 1 : 2$;

d. to apply a temperature of abt 375°C, or with lower concentrations of C_2H_4 at slightly higher temperature.

e. to work with high gas velocity.

* * *

When considering in more detail the characteristic features of the process demonstrated by the S.F.C.G. the following particulars may be mentioned.

1. Composition of the initial gas:

Primarily a mixture of 1 vol. pure ethylene and round 10 vol. air, with the approximate composition of 9 vol.% ethylene, 18 vol.% O_2 and 72 vol.% N_2, was used as base material.

After one cycle a gas of the following composition was obtained, 6,3 vol.% ethylene, 13,6 vol.% O_2, remainder $N_2 + CO_2$. This gas, which gave equally good yields as the primary intake gas, might also be formed as follows:

6,3 vol.% ethylene + 29 vol.% inert gas + 13,6 vol.% O_2 +

51,1 vol.% N_2, i.e.

35,5% ethylene of 18% + 64,7% air, provided that under the reaction conditions the gases accompanying the ethylene are inert in regard to oxygen. In this case even a diluted technical ethylene would, therefore, be suitable.

The intake gas used for the 3rd cycle contained even abt. 3% ethylene on 8% O_2, i.e. abt. 60 vol.% ethylene of 5% on abt. 40 vol.% air.

Further tests would be required to ascertain whether:

a. the CH-compounds accompanying the technical ethylene behave as inert substances during the reaction,

b. the technical ethylene contains catalyst poisons and, if so, how these can be eliminated.

2. *Composition of the catalyst.*

About this no information whatever was supplied.

3. *Reaction temperature and pressure.*

Measured at the tube in the first and second cycles of the gas, the reaction temperature was about 375°C., this being raised in the third cycle to 400—450°C. without giving rise to an abnormal formation of CO_2. *The pressure slightly exceeded atmospheric pressure.*

4. *Addition of water.*

The quantities of water added to the gas mixture to be converted were:

	litres steam of 0°/760 mm
in the first cycle 200 cm³/hr on an average	249
in the second cycle 112 cm³/hr on an average	139
in the third cycle 58 cm³/hr on an average	72
in the cycle on April 8th 1933 233 cm³/hr on an average	289

5. *Quantity of gas led through:*

First cycle 395 l. in 270 min. = 87 l./hr.
Second " 314 l. in 220 " = 86 l./hr.
Third " 232 l. in 160 " = 87 l./hr.
Cycle on April 8th 1933 341 l. in 258 min. = 80 l./hr.

Concentration of ethylene in the mixture of gas and steam passing through the tube.

$$\text{1st cycle } \frac{87}{87 \div 249} \times 9 = 2{,}3\%.$$

2nd cycle $\frac{86}{86 \div 139} \times 6{,}3 = 2{,}4\%$.

3rd cycle $\frac{87}{87 \div 72} \times 3{,}0 = 1{,}7\%$.

cycle on April 8th 1933: $\frac{80}{80 \div 289} \times 9 = 2{,}0\%$.

Ditto concentration of oxygen.

$\frac{87}{336} \times 18{,}8 = 4{,}9\%$

$\frac{86}{225} \times 13{,}8 = 5{,}3\%$

$\frac{87}{159} \times 8{,}6 = 4{,}7\%$

$\frac{80}{369} \times 18{,}6 = 4{,}0\%$

Velocity of gas in the tube:

According to information from the S.F.C.G. the cross sectional area of the quartz tube (22 mm dia.) which unfilled is $\frac{n\,d^2}{4} = 381\ mm^2 = 3{,}81\ cm^2$, has to be reduced to 2,91 cm^2 when taking into account the (unknown) volume of the catalyst. Since, for instance, for the first cycle a total velocity of gas ÷ steam of 336 l. (0°, 760 mm) per hour is calculated from the data of the experiments, i.e. at 375°C.

$\frac{375 \div 273}{273} \times 336$ l. = 810 l./hr., the velocity of the

gas in *cm³ per cm² free section per second* can be calculated as:

$$\frac{810000}{2{,}91 \times 3600} = 77{,}3 \text{ cm}^3/\text{sec.}$$

For the second and third cycles, where less steam is added, this velocity is correspondingly lower and amounts to:

51,8 and 36,6 cm³/sec. respectively.

A few remarks have still to be made in regard to three questions to which no or only little reference has been made in the foregoing, viz.

1. Have the S.F.C.G. already worked out the process so far that it may be applied in practice after a little semi-technical work?
2. Does the process offer advantages as compared with the processes already applied in practice for manufacturing ethylene oxide or glycol from ethylene?
3. What is the characteristic feature of the process and is this feature adequately protected by the S.F.C.G. patents?

With reference to these questions the following is remarked:

1. Since the gentlemen of the S.F.C.G., possibly in view of the fact that the Cie de Béthune intend to exploit their process for France and are already working on semi-technical scale, were unwilling to supply information in regard to tests on a larger scale, only the impression can be given that the following points have still to be worked out:
   a. Can technical ethylene be used without having to eliminate catalyst poisons?
   b. Can technical ethylene be used without considerable concentration? As already remarked, this will principally depend on whether at the reaction temperature the gases accompanying the ethylene are also oxidised in the presence of the S.F.C.G. catalyst. Within certain limits the dilution of the ethylene is of little consequence.

c. Is it possible, by using a well-cooled washing tower, to absorb from the diluted oxide-containing gases the oxide to a reasonably concentrated solution, or is it moreover necessary to work under pressure to achieve this result?

d. Is it possible to make a satisfactory use of the heat of condensation of the large quantities of steam required for carrying out the process?

2. As already remarked, a very accurate calculation will be necessary to ascertain whether the S.F.C.G. process offers advantages as compared with the usual methods. Undoubtedly it has the advantage that no chemicals (chlorine, alkali; etc.) are required, but on the other hand it is possible to convert practically all ethylene into chlorhydrine, which can easily be concentrated, whereas according to the S.F.C.G. process at least 40% of the ethylene is burnt to CO_2, whilst the oxide is primarily present in a highly diluted state. Moreover, in the chlorhydrine synthesis only the ethylene itself is converted, which with oxidation (see question 1 b) is questionable.

3. As a matter of fact this question should be dealt with by the Patent Department and for this reason I will not go far into it. However, it should be stated that in my opinion the only characteristic feature of the process is the *catalyst*, it being possible *to trace all basic characteristics* of the process in the literature on this subject. Consequently it is very important how the S.F.C.G. patents will be worded in countries other than France where a careful examination of patentable matter of the application is made before publication, which will lead to the elimination of the known elements from the matter claimed. Particularly the Dreyfus patent (Brit. Pat. 387,372) runs parallel to the S.F.C.G. patent to such an extent that it is worthy of the greatest attention, although it is stated, probably unjustly, to claim a process for manufacturing glycol.

It might, therefore, be recommendable to negotiate not about patents but about a direct divulgence of the catalyst used, as this seems to be the only valuable element of the process.

DEFENDANT'S EXHIBIT 47

Report on the discussions (on June 17th 1933) concerning the Ethylene oxide process of the Soc. Fr. de Catalyse Généralisée.

The following gentlemen were present:

Mr Masthoff,
Mr Lefort,
Mr Franchon,
Mr Dijt,
Mr Langedijk,
Mr Steenackers,
Mr van Rossum,
Mr Schurink.—

1. Mr Lefort handed Messrs. Masthoff and Dijt a copy of a French translation of the specification of the U.S.A. patent application (No 600,578 dated March 2nd 1932). At our request the original U.S.A. specification will be sent to us.

The first official letter had already been received by the Soc. Fr. from the American Examiner and was said to refer only to formal matter, no anticipating material being cited in regard to the subject matter of the invention. Mr. Franchon remarked that they had been the first to raise and patent the problem of direct manufacture of ethylene oxide from ethylene by oxidation (a provisional research of the respective literature seems to confirm this statement).

Mr Dijt remarked that the examiner frequently first criticizes the form of an application before proceeding to examine the subject matter, so that in the subsequent official letters anticipating material might still be cited. The French gentlemen were quite at ease in this respect.

2. The five French patents (No 729,952 with patents of addition Nos 41255, 41474, 41484, and No 739,562) had been combined to one single application in the U.S.A. In the latter some further details are given concerning a silver catalyst activated with small quantities of gold, copper and iron. This is *not* to be found in the French

applications, although Mr Lefort contended that it is mentioned therein. Later on he stated that it had been taken up in a further patent of addition application (No 30011 dated December 19th 1931) which does not seem to have been published yet; a copy of this application will be sent to us. The American application is worded a little more precisely than the French patent, and Mr. Dijt was of opinion that, if the specification should be accepted with the claims as now on file, the catalyst would be adequately protected provided it falls under some of the claims following on the first claim.

The French gentlemen stated that for *manufacturing* the catalyst (physical state) some special manipulations were required which they were keeping secret, and which consequently were not disclosed in the application. Mr. Dijt remarked that in general this involves a certain risk both for themselves and for their licensees.

The catalyst has a specific effect, only ethylene oxide and carbon dioxide and no other oxidation products being formed.

3. Patents have been applied for in:

France, Germany, U.S.A., Italy, Belgium, Poland, Luxembourg, Roumania, Japan, England and Czecho-Slovakia, and not, therefore, in Holland and Canada where consequently manufacture and import of the product manufactured according to this process are free.

The French gentlemen stated that on Monday, June 19th, they would be leaving for Frankfurt to visit the I.G., who intend to acquire the rights of the whole of Europe excl. France and Italy.

In France the Cie des Mines de Béthune is investigating the process on a technical scale, whilst in Italy the Government was said to be interested. Further they wished to reserve Japan. It was stated that the I.G. wished to acquire the patent in order to apply it themselves and not only in order to prevent the exploitation by third parties.

It was remarked that the process was not patented in the British colonies and Dominions.

The French gentlemen were informed that the Group would be principally interested in the process for U.S.A.

4. In the U.S.A. we have available two sources of ethylene:

a. A fraction obtained when purifying the hydrogen for the ammonia process, which contains about 50% ethylene (45.9% C_2H_4, 23.4% CH_4, 30.7% C_2H_6).

b. Crack-gases, which, however, contain only 5 to 10% ethylene (sometimes even less).

Mr Franchon was of opinion that the first gas could be worked without difficulty; he was not so sure about the second gas, a content of 40-50% ethylene being required (according to him).

Dr Langedijk remarked that the gas re-circulated in the second and third cycles of the tests made in his presence at Paris had much smaller contents of ethylene and that nevertheless satisfactory yields (calculated on converted ethylene) had been obtained.

Mr. Franchon contended that in order to obtain a satisfactory yield the following combination of proportions was required in the reaction gas (in which combination no physical or chemical signification whatever can be found):

$$\frac{O_2}{C_2H_4} \times \frac{O_2 + C_2H_4}{\text{Total diluents}} = \text{abt. } 0{,}8 - 1,$$

With reference to figures from the tests made at Paris Dr Langedijk proved that this could not be true, since for the respective gas this "factor" was only 0,4 — 0,5. Mr Franchon then admitted that the figures cited by Dr Langedijk indeed showed that these values might suffice. The French Coke Oven Works obtain a coke furnace gas containing abt 2% ethylene, which is concentrated to 50%, which is said not to be very expensive.

It has still to be investigated whether crack gases containing also other unsaturated components are equally suitable for the reaction and whether the ethylene can be equally well concentrated.

5. It was discussed what further steps would now have to be taken. We stated that the Shell Development Comp. intended to examine the process in their laboratories themselves and requested the French gentlemen to inform us about the composition of the catalyst and supply further data required for carrying out the tests without difficulties.

However, the gentlemen did not care to do so, as according to their statements the suitability of the catalyst depends on a few special manipulations during manufacture which are not described in the patents and which they were unwilling to divulge to us before an agreement had been made.

We then requested them to despatch to the Shell Development Comp. the ready catalyst. This they were prepared to do, but they asked for a sum of Frs 500,000.—, on payment of which we would be entitled to experiment with their catalyst for six months. At the expiration of that period the money would either go to them in case the matter fell through or be settled with the payments to be made in the event of a license agreement being entered into.

We did not agree to this proposal, and suggested to ask the Shell Development Comp. to send samples of their gases to Europe in order to have tests made with them in Paris by the "Catalyse" under the supervision of one of our chemists. These tests might then be repeated at our Amsterdam laboratory in the presence of one of the gentlemen of the "Catalyse". It was agreed that the running of such tests would not bind us in any way towards the Soc. de Catalyse Généralisée.

In order to be able to submit more or less concrete proposals to the Shell Development Comp. the French gentlemen were requested to inform us about their conditions for

a. an exclusive license for the U.S.A.

b. an exclusive license for the U.S.A. plus the right of free sale over the whole world, with the exception of Europe and Japan, of the ethylene oxide and glycol manufactured in U.S.A. by the Group.

Within eight days Mr Lefort would confirm in writing the provisional arrangements made and name their conditions.

It was agreed that we should inform the Shell Development Comp. about the French process, the discussions held and the conditions of the Soc. de C.G.

Should the Shell Development Comp. after studying this information still be interested in the process, then the tests with their gases mentioned above would be carried out.

In order not to lose any time Shell Development Comp. would be asked to have the gases in question shipped at once. As it would take at least two months before the tests could be started, there would be ample opportunity in the meantime to study Dr Langedijk's report on the process and the patents.

Further it was discussed what time would be required for making the necessary arrangements and carrying out the tests. As it was calculated that in all about six months would be involved, we have been granted an option for six months until Dec. 31st 1933 without any commitment or payment by us.

Mr Lefort remarked that as a matter of fact we had now already obtained something from them, viz. the promise that in the meantime they would not negotiate with third parties, and that this promise was also worth something. This point was not discussed further, since —as was remarked on our side—it was in the interest of both parties to have the tests carried out.

6. Information was asked for about the cost price of the catalyst in order to enable us to determine the capital to be invested. On this point the French gentlemen were very reserved and stated that the catalyst, just as gold and silver, retained its value. Neither did they inform us about the life of the catalyst, which they stated to be very long, whilst the catalyst could easily be regenerated. The catalyst poisons are sulphur compounds (also arsenic).

7. Mr. Franchon remarked that glycol from ethylene according to their process could be produced in Europe

at a costprice of at most Frs. 3.—per kilo, incl. capital service, with an ethylene price of Frs. 0.66 per m^3 (calculated on 100%). The present selling price in America was stated to be Frs. 11.- — 14.- per kilo.

8. In reply to a question of Mr. Masthoff in regard to the data available for application of the process on a large technical scale, Mr Franchon stated that complete data for equipment etc. had been worked out by competent engineers of Construction companies and that these would be put at our disposal as soon as a contract would be closed.

DEFENDANT'S EXHIBIT 48

E. L. MASTHOFF

'S-GRAVENHAGE, July 6th, 1933.
CAREL VAN BYLANDTLAAN 30

—. BY AIRMAIL .—
D. PIJZEL, Esq.,
Shell Development Company,
100, Bush Street,
SAN FRANCISCO (Cal.)
U.S.A.

CI (CC-OA)—Société Française de Catalyse Généralisée

July 6, 1933

My dear Pijzel,

Further to my letter N°555 of the 28th June I would inform you as follows:

We received a letter from the Société Française de Catalyse Généralisée dated 28th June, of which you will find a copy enclosed.

On comparing the contents of this letter with the minutes of the conference sent to you with my above-men-

tioned letter you will notice that the people of the Société have introduced conditions which are totally in contradiction to what was agreed upon at The Hague. They now want a complete contract signed by both parties, to become effective if the results of the tests with our ethylene gas are in conformity with the tests made in March last, described in Dr Langedijk's report, and if we feel satisfied about the patent position.

From the minutes it is clear that no such thing has been discussed at all, and that it was agreed that neither the option of 6 months nor the results of tests with our ethylene, nor anything else would bind us.

You will also find enclosed copy of our reply of the 5th July to their letter, to which we added that we think their financial conditions too excessive to be discussible.

It is, of course, difficult for us on this side to estimate the value of their process for U.S.A. from the standpoint of commercial application, and you will be in a better position to judge this, but if I may venture an evaluation and take it that the patent position etc. is all right, I should think that a tenth of the cash payment asked for is about what we could consider plus a royalty of say maximum 0.5 French francs per kg ethylene oxide produced as such or in other products. I just heard from Riemersma of Hercules Powder Co. that they buy ethylene glycol at ¢ 12/lb for 10 ton lots. This is ¢ 26.4 per kg, and considering a costprice in U.S.A. of Fr.4.50/kg or ¢ 18 (all in dollar at par), instead of Fr. 3.—, I feel that Fr.0.5 or ¢ 2 royalty per kg oxide would be the utmost to go to.

However, it is for you to consider these questions and give us your advice. In the meantime we shall abide by what has been agreed upon in our conference of June 17th and if they agree and the tests with our base materials plus patent position have been found satisfactory, then I think the Société will have to furnish us with their complete data on equipment required, its price and operating costs before we discuss the financial conditions of the proposed deal.

Enclosed you will also find the note from our OA department on the patent side of the French process referred to in my letter of the 28th June.

As regards Hercules Powder Co.'s interest in this process, I heard from Riemersma that his directors do not consider to buy it.

Yours very truly,

E. L. Masthoff.

encl.:

copy of letter from S.F.C.G. dated 28th June,
" " " to S.F.C.G. " 5th July,
" " memo from OA Dept.

c. c. Mr. C. B. deBruyn.

DEFENDANT'S EXHIBIT 49

Translation

Société Française de Catalyse Généralisée

Paris, June 28, 1933

N. V. deBataafsche Petroleum Maatschappij
Carel v. Bylandtlaan 30
La Haye
Holland

Gentlemen:

First allow us to thank you for the excellent reception accorded to our agents during the call they made on you.

We are today in receipt of your letter of the 27th inst., and beg to be excused for not having written you earlier but our board of directors met only yesterday.

We are sending you enclosed herewith a copy of our U.S. patent in accordance with your desire.

On the other hand, it has been agreed upon that we shall repeat in our Courbevoie laboratory the experiments which you have witnessed some time ago and the result

of which has been recorded in a report dated March 31, with gases derived from sources of your own.

If you so desire these experiments will likewise be repeated in your Amsterdam laboratories. It is therefore important that you send these gases promptly to us by the expeditious route.

In short we agree in principle subject to the conditions set forth above to grant you a license in North America and facilities for selling products manufactured in Canada and the Dutch East Indies.

You asked us to indicate to you the main conditions of this grant.

The license rights would be acquired by you on payment, in cash, of the sum of frs. 2,000,000. (two million francs), and of a royalty of one franc per kg. of manufactured products.

Kindly advise us whether you agree on these bases; in the affirmative case we shall have a draft of the complete contract drawn up so as to put us in a position where we can exchange our signatures contingent on the successful outcome of the experiments.

In case the results of the experiments are consistent with the report of March 31, 1933 and you are satisfied with respect to the question of the patent the contract would ipso facto become final.

This signature would on our part be worth an agreement not to cede this right to anybody for the United States until the termination of the experiments.

As soon as your investigation of the patent is finished we would ask you to let us know your conclusions with regard thereto.

We are awaiting your reply and, as the case may be, your objections and ask you, Gentlemen, to accept the assurance of our most distinguished sentiments.

The Delegate Director
General Manager

(signature illegible)

Translation.

To

Société Française de Catalyse Généralisée
8 Avenue Percier
Paris (VIIIe)

La Haye, July 5, 1933

CC (CI-OA)

Gentlemen:

We acknowledge receipt of your letter of June 28 stating your conditions for a license covering the application of your process for the production of ethylene oxide, and also receipt of the copy of your American patent which copy has been sent, as agreed upon, with all the necessary details to our friends in California.

With respect to your confirmation of the agreement entered into with your Messrs. Franchon and Lefort we much regret having to observe that the confirmation is not in accordance with what had been agreed upon. This difference we presume must be attributed to a misunderstanding on your part.

In order to clear up this matter we are sending you enclosed herewith for your guidance a memorandum (in English with translation into French) drawn up after the discussions we had with your representatives on Saturday June 17, paragraph V of which precisely sets forth the agreements entered into. We shall appreciate receiving confirmation from you to the effect that you agree with the contents of this memorandum.

Concerning the cash sum of frs. 2,000,000.—and the royalty of 1 franc per kilogram of manufactured products which you ask, we feel bound to tell you that we are unable to take into consideration any payment of such magnitude.

If there is on your part any incentive to have an interview with us in order to clear up the misunderstanding we are entirely agreeable to receive your representatives.

In the expectation of hearing from you we are sending you our fervent greetings.

N. V. deBataafsche Petroleum Mij. (in pencil)

signature H. Jacobson (stamp)

Enclosures

DEFENDANT'S EXHIBIT 50

H. JACOBSON

's-Gravenhage, 17th July, 1933.
Carel van Bylandtlaan 30

VIA AIR MAIL.
D. Pyzel, Esq.,
100, Bush Street,
SAN FRANCISCO.

CC(CI) Ethylene-oxide.

July 17, 1933

My dear Pyzel,

Please find enclosed for your information copy of a letter from Société Française de Catalyse Généralisée, Paris, on the above subject, together with copy of our today's reply.

As you will see, the French gentlemen wish to fix the conditions for a licence before the 31st August 1933 and I should appreciate it very much if you could see your way to let us have your opinion on the process and its possibilities, together with your idea of the price we should pay, in the second half of August.

Thanking you in advance for the trouble to be taken,

Yours sincerely,

H. JACOBSON

Encs. 2 copies

DEFENDANT'S EXHIBIT 51

Translation.

Société Française de Catalyse Généralisée
8 avenue Percier
Paris (8e)

Paris, July 13, 1933

To

N. V. deBataafsche Petroleum Mij.
Carel v. Bylandtlaan 30
La Haye
Holland

V/R CC(CI-OA)

Gentlemen:

We have the honor to acknowledge receipt of your letter of the 5th inst.

Enclosed herewith please find a copy of the addition No. PV 30011 to patent No. 739562 dated Oct. 3, 1931.

In principle we are in accord with the terms of the report attached to your letter.

As concerns the option period we are likewise in accord in that you hold an option until Dec. 31st. It is however our formal desire, in order that this option period be granted you in a final manner, that we come previously to an agreement with respect to the financial conditions under which you will buy our process from us in case of the successful issue of the tests.

You must understand that it would be useless for us to bind ourselves for six months if we are never to come to an agreement with respect to the financial conditions.

It is therefore necessary for us to make provision for these conditions beforehand in the event of the successful issue of the tests. It seems to us that this agreement could be reached before Aug. 31 next.

For the present we give you until December 31 to make a decision. From now until such time as this agreement comes into force—and we think that this can be done by Aug. 31—we shall enter into no negotiations with other parties.

As concerns these conditions you tell us that they are too high. We are asking you therefore to make us counter-proposals with respect thereto.

We are entirely at your disposal for a return journey to La Haye and for renewed conferences with you on this subject, from Monday July 24 on.

Please accept, Gentlemen, the assurance of our most distinguished sentiments.

(signed) Lefort

Translation.

To
Société Francaise de Catalyse Généralisée
8 avenue Percier
Paris (VIIIe)

La Haye, July 17, 1933

CC(CI-OA)

Gentlemen:

We are in receipt of your letter of July 13 from which we gather that in principle you agree with us as to the wording of our report covering the discussions held in our office on Saturday, June 17.

With respect to the financial conditions for a contingent license on our process we are quite ready to negotiate with you with a view to fixing these conditions in case we should decide to acquire your rights for the United States. The Shell Development Co. of San Francisco who are interested in your process and for whom we act as go-betweens are now doing research work on the possibilities of your process and the products in the United States.

You understand that it is necessary to wait for the report before we are in a position to resume the negotiations with you. We asked our friends in California to send us the necessary informations as soon as possible, and hope to be able to resume the negotiations at the end of the month of August.

We are willing to believe that in the meantime you shall not enter into negotiations with other companies, and shall appreciate it if you can let us have your confirmation.

Please accept, Gentlemen, the assurance of our most distinguished sentiments.

N. V. DeBataafsche Petroleum Maatschappij.

DEFENDANT'S EXHIBIT 52

H. Jacobson

's-GRAVENHAGE, 25TH JULY, 1933
CAREL VAN BYLANDTLAAN 30

VIA AIR MAIL.
D. PYZEL, Esq.,
100, Bush Street,
San Francisco.

CC (CI) Ethylene-oxide.

July 25, 1933

Dear Mr. Pyzel,

Further to our letter of 17th July on the above subject I have pleasure in enclosing for your information copy of a letter from Société Française de Catalyse Généralisée, Paris, to Bataafsche, dated 20th July.

Yours sincerely,

for H. Jacobson
Van Rossum.

Enc. 1.

DEFENDANT'S EXHIBIT 53

Translation.

Société Française de Catalyse Généralisée
8 avenue Percier
Paris (VIIIe)
Paris, July 20, 1933

To N. V. DeBataafsche Petroleum Mij.
Carel v. Bylandtlaan 30
La Haye

Gentlemen:

We beg to acknowledge receipt of your letter of the 17 inst.

We shall therefore expect to hear from you before Aug. 31. From now until then we shall enter into no negotiations with another company as far as the United States are concerned.

We think it would help matters if the gases which your friends in California are to send, arrive here as soon as possible.

In case you need us for any reason whatever we are entirely at your disposal.

Please accept, Gentlemen, our most distinguished salutations.

(signed) Lefort.

DEFENDANT'S EXHIBIT 54

Report No. 1759

SHELL DEVELOPMENT COMPANY
EMERYVILLE

MONTHLY PROGRESS REPORT
AUGUST, 1933

By

H. Groll.

SUBJECTS: 708. Removal of Mercaptans from Olefines. (H. Groll and G. Hearne).

718. Glycols, Oxides and Related Substances. (H. Groll and H. de Jong).

1314. Isobutyric Acid and Allied Products. (H. Groll, G. Hearne, C.J. Ott and H. de Jong).

For the information of the Director of Laboratories only. Not for distribution.

No. 1759

718. Glycols, Oxides and Related Substances.
(*H. Groll and H. de Jong*).

All experiments on production of ethylene oxide from ethylene and air under conditions intended to imitate the Société Française de Catalyse Généralisée's process were futile. However, this by no means disproves the claims of the Société Française de Catalyse Généralisée. We could not find a catalyst of the right activity. Most of our catalysts did not oxidize the ethylene at all. Hopcalite however catalyzes complete oxidation to CO_2 and water even at 275°C. So it appears that there should exist some combination of catalyst which has just the right degree of activity. A detailed report on this is being prepared.

H. Groll

/IF
August, 1933

DEFENDANT'S EXHIBIT 58

/ Sixth
October
1933

D #439

De Bataafsche Petroleum Mij.,
30 Carel van Bylandtlaan,
The Hague, Holland.

Catalyst Composition: S.F.C.G. Process

Gentlemen:

I am herewith sending you the desired information regarding the approximate composition and method of preparation of catalysts for the ethylene oxide process of the S.F.C.G., which you requested in your cables No. 300 of October 2nd, and No. 853 of October 5th:

Catalyst #1

Pure silver was turned on the lathe to form small shiny turnings.

Catalyst #2

Silver turnings were placed in a dish and very clean mercury sprayed over it through a chamois bag with good stirring. The mixture was placed in a glass funnel, in order to drain off the excess mercury, and allowed to stand for about three days. At the end of this time, the mercury was removed under reduced pressure at 500°C.

Catalyst #3

100 parts of silver turnings and 5 parts of powdered copper (Naturkupfer C) were mixed in a flask. To this, 35 parts of mercury were added slowly while the flask was shaken vigorously. When all mercury had been added, about 8 parts of very dilute HNO_3 was added and the shaking continued for 10 minutes. The whole was then freed from water and the mercury removed under reduced pressure at 500°C.

Catalyst #4

Finely powdered metallic arsenic was mixed with silver turnings and heated quickly in a covered crucible to give an alloy rich in arsenic. This alloy was placed on the bottom of a graphite crucible and molten silver poured on top of it. The resulting block of alloy was melted again, covered with charcoal, stirred well, and poured into an iron mold. The arsenic content was about 4%.

The rod cast by this method was turned on the lathe and the turnings used as catalyst without further treatment.

Catalyst #5

1 part of tin was dissolved in very dilute HNO_3 and enough water added so that 60 parts of water were present. To this was added 1 part of each bismuth, silver, lead and nickel, as nitrates. When all were dissolved, 100 parts of 8-10 mesh pumice were added with good stirring.

The water was then evaporated off and the catalyst roasted until no more brown vapors were given off.

Catalyst #6

1 part of antimony, as Sb_2O_3, was melted on 100 parts of pumice in an electric oven. The catalyst was then prepared with the same constituents and in the same manner as Catalyst #5, but before roasting 1 part of arsenic was added as H_3AsO_4.

Catalyst #7

Same as #6, but without bismuth.

Catalyst #8

Same as #6, but without lead.

Catalyst #9

Same as #6, but without lead and tin.

Catalyst #10

Same as #6, but without antimony and bismuth.

Catalyst #11

Same as #6, but without antimony and lead.

Catalyst #12

So far only tried in unreduced state—"Hopcalite", as supplied by the Mine Safety Appliances Co. at Pittsburg, Pa., for use in gas masks.

Yours very truly,

SHELL CHEMICAL COMPANY
Original Signed by D. Pyzel

DP:B

DEFENDANT'S EXHIBIT 59

E. L. MASTHOFF

's-GRAVENHAGE, 2nd October, 1933.
CAREL VAN BYLANDTLAAN 30

BY AIRMAIL.
D. PIJZEL, ESQ.,
c/o. Shell Development Co.,
100, Bush Street,
SAN FRANCISCO.
10/2/33

CI (CC-OA)

My dear Pijzel

From your cable No 154 of the 28th September I duly noted that the Emeryville experiments re the ethylene-oxide process of the Soc. de Catalyse Généralisée were completely negative and that you could not give us your recommendations which would put us in a position to make a counter-proposal to the Soc. de Cat. Gén.

With my letters of the 28th June and 6th July I submitted to you all information available re the French process with the idea that Shell Development would study the process from this information and compare it with the chlorhydrine process with which Shell Development are experimenting. That Shell Development should make their own experiments on the French process did not come into consideration, because it was believed that satisfactory results could not be obtained without knowing the exact composition of the French catalyst. This now seems to be substantiated by your cable No 154.

Of course, a comparison as mentioned above—for which certain yields in the French process would have to be taken as a basis, e.g. 40 and 45% yield as oxide of the ethylene introduced—could not claim to be exact. It would as a matter of fact be principally a comparison on a basis of the cost of ethylene and chemicals consumed, but I thought that the result might in the first place enable Shell Development to express an opinion as to whether the French process had sufficient attractiveness

above the chlorhydrine process to warrant further negotiations on the acquisition of the U.S.A. rights, and the hope that the outcome of this comparison and a study of the matter from a marketing point of view, would enable you to express an opinion as to what would be a reasonable price to pay.

The experiments with ethylene containing gases of the Group's produce, either directly available as in the case of the Shell Point ethylene fraction or which could be made available from Dubbs gas or by special cracking of certain base materials, which were envisaged in co-operation with the Soc. de Cat. Gén., would only be taken after a favourable opinion on the French process had been expressed by Shell Development Co., whereas the results of these experiments would then serve as the deciding proofs with regard to the profitability of the French process.

I will, of course, discuss with you and Dr Williams this question during my forthcoming visit and I should very much appreciate it if in the meantime Shell Development would study the matter from the viewpoint as explained above, so that we may jointly give our recommendation to The Hague as soon as possible.

As already said above, the completely negative results of Emeryville's experiments seem to confirm our opinion that the patent applications disclose very little indeed of the essential part of the invention, viz. the catalysts required.

With their letter of 12th September, copy of which is enclosed, the Soc. de Cat. Gén. informed us that experiments based on the information in our possession, will not give satisfactory results and that additional information *not disclosed in the patents* is required, which secret information can only be provided by them.

It seems a rather imprudent move on the part of the Soc. de Cat. Gén. to make such a written statement, by which they would themselves confirm that their patent applications when attacked run the risk of being declared invalid on account of insufficient disclosure, whereas any outsider might get patent protection on a catalyst developed along the general information in the Soc. application, when this outsider claims a detailed description of such catalyst.

In view of this, we had, therefore, already decided to carry out experiments in the Amsterdam Laboratory on the French process, in the first place to satisfy ourselves that their statement is right that we cannot get satisfactory results when using the information contained in their patents only, and further to try to find ourselves those conditions by which a suitable catalyst can be developed.

Of course, you will be kept "au fait" of any findings by the Amsterdam Laboratory.

Yours sincerely,

E. L. MASTHOFF

encl.:
copy of letter from Soc. de Cat.
Gén. dated 12th Sept.

c.c. *Mr. De Bruyn.*

DEFENDANT'S EXHIBIT 60

Translation

SOCIETE FRANCAISE DE CATALYSE GENERALISEE

PARIS, le 12 Septembre 1933.

N. V. DE BATAAFSCHE PETROLEUM MIJ.,

LA HAYE.

CC(CI-OA)—

Gentlemen:

We have received your letter of September 4.

Before giving you a time option, which you have requested, until December 31 for example, we would like to be certain about several points.

In fact we do not understand why your friends in California have not been able, in all this time, to make a decision.

You tell us that they are doing research which will require still more time; that is what we do not understand, and we would like to know why their research takes such a long time. What research can they be doing?

Either this research concerns our patents, and in this case your California friends should have made up their minds. Or this research is of a technical nature, your friends for example are doing experiments; and if that is the case, we are then obliged to tell you that with the data they have on hand they will not be able to obtain satisfactory results.

Without complementary explanations from us as to the tricks of the catalyst, any experiments which they make can only be the cause of considerable loss of time.

It is for this reason that we do not see the necessity of waiting until the end of December.

Besides, you inform us that you will eventually be interested in Italy. Now, we are very much afraid, in view of the time which your California friends require for their decision, that we will have made an agreement in Italy before their response about America reaches us.

We will be very much obliged if you will be good enough to advise us regarding these various points, so that we can make a decision.

We would like to hear from you very soon, and await your reply we send you our good wishes.

l'Administrateur-Délégué
Directeur Général

DEFENDANT'S EXHIBIT 61
Nineteenth
October
1933

D #474

De Bataafsche Petroleum Mij.,
30 Carel van Bylandtlaan,
The Hague, Holland.

CI (CC-OA)

Re: Ethylene Oxide Process of the
S. F. C. G.

Gentlemen:

I refer to Mr. Masthoff's letter No. 580, of October 2nd, dealing with the ethylene-oxide process of the Soc. de Catalyse Generalisee.

I will discuss the contents of this letter with Mr. Masthoff when I meet him in Chicago next week, but in the meantime, I want to draw your attention to the fact that the object of the experiments carried out at the laboratory in Emeryville was to determine whether or not the disclosures made in the patent applications of the S. F. C.G. were sufficient to enable "anybody versed in the art" to carry out the process as is required, in order to make a U. S. patent valid. Apparently this is not so and I see from the second paragraph on page three of Mr. Masthoff's letter that the S.F.C.G. admit that the necessary information is not disclosed in the patents.

Under these circumstances these patents would be held invalid in this country, which means, of course, that they would have very little value for us. As I cabled you on August 29th, I consider that the royalty of 1 franc per kg. of ethylene-oxide is far too high, while a cash payment of two million francs would be out of the question.

Yours very truly,

SHELL DEVELOPMENT COMPANY

Original Signed by D. Pyzel

DP:B

DEFENDANT'S EXHIBIT 64

November 3, 1933

AIR MAIL

MR. H. JACOBSON
De Bataafsche Petroleum Mij.
30 Carel van Bylandtlaan
The Hague, Holland

Re: Ethylene Oxide/Glycol

My dear Jacobson:

Upon my arrival here in the beginning of this week, I found Hardonk's letter on the recent discussions with S.F.C.G. at the Hague, and I cabled yesterday Pyzel's and my comments as asked for.

You will see from this cable that we consider the scheme for further investigation of S.F.C.G.'s process and negotiations with them, as set forth in the resume of the discussions, to be satisfactory if revised in such way that cost prices derived from laboratory experiments only, are entirely disregarded as a basis for financial obligations on Bataafsche's part. * * *

If they would keep on insisting on a basis of laboratory calculations, I should say that we then better leave them alone. It should not be overlooked that their patent situation seems rather insecure for lack of sufficient disclosure in their claims, so that if somebody also gets out a patent on a specified catalyst, we might find ourselves paying royalties for a process which others can apply without such payments. The chance that this happens should not be neglected for extensive research work on hydrocarbons is going on everywhere. * * *

Yours sincerely,

E. L. MASTHOFF

ELM:MD

DEFENDANT'S EXHIBIT 65

SHELL DEVELOPMENT COMPANY

SIXTH REPORT

PERIOD ENDING DECEMBER 31, 1933

718. GLYCOLS, OXIDES AND RELATED SUBSTANCES (*H. Groll, G. Hearne, C.J. Ott and H. de Jong*).

Purpose.

This investigation was opened during the period under review mainly in order to segregate various activities formerly dealt with under Investigations 717, 1313 and 1314. * * * Also, a limited investigation of the direct formation of ethylene oxide from ethylene and air according to the S.F.C.G. process was undertaken with the aim of checking the patent claims of the French company.

The different subjects are closely related by the character of the products as well as the methods applied and can be subdivided as follows: * * *

(3) Production of ethylene oxide from ethylene and air (S.F.C.G. process).

Programme. * * *

(3) Ethylene and air in the proportions recommended in Dr. Langedijk's report (B.P.M. No. 5205) were passed over a large number of catalysts in an attempt to reproduce the S.F.C.G. process with the data available from the patent specification.

Summary. * * *

(3) Direct Production of Ethylene Oxide from Ethylene and Air—S.F.C.G. Process.

In connection with the discussions in progress between The Hague and the Société Française de Catalyse Généralisée relative to the securing of rights to the latter process for making ethylene oxide by the direct catalytic

oxidation of ethylene, we have carried out experiments to verify the claims of the relevant patents.

It has been extremely difficult to carry out the above programme at Emeryville because the S.F.C.G. refuse any information on the nature of their catalyst. This is, of course, the essential feature of the process.

We were therefore left with the unsatisfactory course of trying as many varieties of catalysts as possible in the time available, being guided in our choice of catalysts by the inadequate disclosures of the S.F.C.G. patents. If this course led to successful results it would at least prove reasonableness in the claims made by the S.F.C.G.; if it led to unsuccessful results it would, however, not disprove the claims but only mean that we had not found such a good catalyst as the S.F.C.G. claim to have. It is axiomatic that the activity of a contact catalyst is dependent upon its history about as much as upon its chemical constitution.

The S.F.C.G. patents, which were our only basis for the investigation, claim the use of a large variety of metals and metal oxides, used either singly or in pairs or larger groups. The metals actually covered by the S.F.C.G. are silver, copper, gold, arsenic, antimony, tin, iron, bismuth, lead, and nickel, whence it will easily be seen that the possible permutations and combinations are enormous, especially if we consider the different forms in which the ingredients may be prepared.

We have tried in our experiments silver turnings, copper turnings, silver activated by amalgamation and heating (known to give a good active silver for dehydrogenations); silver and copper alloyed together by amalgamation and heating; also a complex mixture, as described in the S.F.C.G. American patent application, of antimony, silver, lead, arsenic, nickel, and tin, impregnated on pumice and used in both reduced and unreduced forms; also mixtures containing part of these ingredients in which single elements or pairs were omitted, according to a regular scheme.

In all the experiments with the above catalysts we obtained practically no ethylene oxide at the temperatures recommended by the S.F.C.G. and practically no reaction at all until the temperature was raised to about 500°C.,

at which point a little oxide was obtained but was invariably accompanied by aldehydes, in contradiction to the statements of the S.F.C.G., which claim the exclusive production of ethylene oxide free from aldehydes.

Steam was used in all cases in the preparations as recommended in Dr. Langedijk's report, B.P.M. No. 5205.

In short, we have been unable to get any appreciable oxidation of ethylene with catalysts prepared to the best of our skill and under the guidance of the S.F.C.G. patents and the description of the process as furnished in B.P.M. Report No. 5205. This does not mean that we disagree with the claims made in these patents but only that the disclosures available to us have not enabled us as yet to confirm them. In all the above experiments we started at temperatures around 375°C., as recommended in the B.P.M. report, and after verifying that no reaction occurred there, slowly raised the temperature, still looking for reaction, up to 500°C. It was at this higher temperature, as a rule, that reaction started but with the formation of only traces of ethylene oxide and that accompanied by aldehydes.

In order to determine whether even more powerful though not recommended catalysts were active at 375°C., we tried as an extreme measure the catalyst hopcalite (a mixture of the oxides of the elements copper, cobalt, manganese and silver) in both reduced and unreduced forms.

With this extremely active catalyst in the oxidized form, we obtained oxidation of ethylene below 275°C. This temperature was the highest which could be maintained without the catalyst beginning to glow. The products at all temperatures between 150 and 275°C. were solely CO_2 and H_2O.

After reducing this catalyst with hydrogen at 350°C. it was tried out between 250° and 400°C. At 250°C. the ethylene was not attacked at all, while the oxygen disappeared from the gas due to oxidation of certain constituents of the catalyst. At temperatures toward 400°C. some ethylene was consumed and a corresponding amount of CO_2 formed. No trace of ethylene oxide could be found at any temperature.

Our present position with regard to this investigation is therefore that while ethylene can be oxidized with an extremely active catalyst at temperatures as low as those recommended by the S.F.C.G., only CO_2 and H_2O are formed thereby, and while we may succeed in finding a catalyst of lesser activity which will permit controlled oxidation to ethylene oxide, we have so far been totally unsuccessful in finding it.

Assessment. • • •

(3) The process of the S.F.C.G. for production of ethylene oxide could not be reproduced. It appears therefore that the essential features of this process are not sufficiently disclosed in the patent application of that firm. • • •

UNITED STATES PATENT OFFICE

2,040,782

MANUFACTURE OF OLEFINE OXIDES

Adrianus Johannes van Peski, Amsterdam, Netherlands, assignor to Shell Development Company, San Francisco, Calif., a corporation of Delaware

No Drawing. Application November 5, 1934, Serial No. 751,579. In the Netherlands April 26, 1934

23 Claims. (Cl. 260—54)

This invention relates to a novel process for the manufacture of olefine oxides which comprises reacting an olefine with free oxygen, which oxygen may be present as oxygen per se or in admixture with other gaseous materials as steam, nitrogen, carbon dioxide, etc., said oxidation being effected in the presence of an active catalyst substantially comprising silver metal prepared by effecting the thermal decomposition of an organic silver compound.

An object of my invention is to provide a practical, economical and broadly applicable method for effecting the catalytic oxidation of olefines to the corresponding valuable olefine oxides.

The olefine oxides are valuable compounds per se as solvents, extractants, fumigants and insecticides and the like. In addition, they are valuable intermediates in the preparation of a wide variety of useful organic compounds such as alcohols, glycols, aldehydes, carboxylic acids, ethers, esters, alkylolamines, resins, polymers and the like.

Heretofore no practical method has been proposed for the production of olefine oxides by effecting the direct oxidation of the corresponding olefines. It is known that ethylene may be reacted with oxygen to obtain small amounts of ethylene oxide and large quantities of formaldehyde, and carbon oxides. Various catalysts and methods of controlling the oxidation to increase the yields of ethylene oxide have been proposed.

I have attempted to prepare ethylene oxide in accordance with the disclosed methods. These attempts were unsuccessful. Under optimum conditions of operation, in accordance with the known methods, employing as catalysts silver per se, colloidal silver and silver prepared by effecting the decomposition or reduction of inorganic silver compounds, only traces of ethylene oxide were obtained, while substantially complete oxidation of the ethylene to carbon dioxide occurred. Due to the low yields of ethylene oxide obtained and the substantially complete conversion of the ethylene to carbon dioxide, the known methods of oxidizing ethylene have failed to warrant consideration as practical processes for the technical production of ethylene oxide.

Now I have found that, in the presence of a particularly active catalyst which I prepare by effecting the thermal decomposition of an organic silver compound, I may oxidize an olefine to its corresponding olefine oxide in yields sufficiently high to render my process commercially attractive as a method for the manufacture of olefine oxides.

In the execution of my invention I preferably employ the normally gaseous or readily volatile unsaturated organic compounds such as ethylene, propylene, the butylenes, the amylenes and their homologues and suitable substitution products. I may employ the olefines severally or in mixtures containing a plurality of different species of olefines, or I may resort to the use of mixtures thereof with relatively unreactive substances. For example, mixtures of olefines and paraffins may be employed without resorting to the separation of the olefine or olefines therefrom. Such olefine-containing mixtures may be obtained by the pyrogenesis or cracking of mineral oils such as petroleum oil and shale oil, by the destructive distillation of various kinds of brown coal, by the cracking of natural carbonaceous material as petroleum products, tars, pitches, asphalts and the like.

In addition to the above-described mixtures of olefines with relatively inactive organic compounds, I may employ an olefine or olefines in admixture with inactive gases such as carbon-dioxide, hydrogen, nitrogen, steam and the like.

The successful execution of my invention is dependent on the use of active silver catalytic material which is obtained by effecting the thermal decomposition of suitable organic silver compounds. Suitable organic silver compounds, in general, are those which, on being heated, will decompose with the formation of an active and finely divided silver catalytic material. The silver salts of the carboxylic acids such as formic, acetic, propionic, butyric, isobutyric, valeric, oxalic, malic, molonic, lactic, malaic and the like are particularly suitable organic silver compounds which may be readily decomposed to catalytic silver by heating the same at temperatures at which decomposition can occur. I have found that silver oxalate is a particularly suitable organic silver compound which I may readily decompose with heat to form an active silver catalytic material. Silver oxalate is practically insoluble in water. Accordingly, it may be readily precipitated by the addition of a solution of silver nitrate to a solution of an oxalate. The suitable organic silver compounds may be decomposed in an atmosphere of hydrogen or other suitable reducing gases or decomposition may be effected in the presence of an inert gaseous substance such as nitrogen. In some cases a more efficacious catalytic material may be prepared by dissolving or suspending a suitable organic silver compound in a suitable inert liquid such as petroleum oil, hydrocarbon and the like and the mixture heated to the decomposition temperature of the silver compound in the presence or absence of hydrogen or other reducing gases.

The catalytic effect of the silver catalytic material prepared and used in accordance with my invention may be considerably enhanced by the admixture with the catalytic material of small quantities of other materials capable of acting as promoters. Suitable promoters, which may be used singly or in combination, include the metals such as copper, gold, iron, manganese, nickel, cobalt, cerium, thorium and zinc. I have found that gold and manganese are particularly effective promoters. A suitable promoter or promoters may be incorporated with the mass of silver catalyst in any suitable manner. Preferably, an intimate mixture of the organic compounds of silver and the promoter metal is effected by some suitable means as by precipitation and the same decomposed by heating.

In general, the catalytic material is quite voluminous and in a finely divided state, accordingly, the same may be advantageously employed without resorting to the use of a carrier. However, when desired, the catalytic material may be deposited upon an inert substance or carrier such as pumice, charcoal, activated carbon, silica gel and the like.

My invention is executed by reacting an olefine with free oxygen in the presence of a suitable catalyst as herein described and preferably the same is executed in the presence of a relatively inert gaseous substance which acts as a diluent such as steam, carbon dioxide, nitrogen, a paraffin hydrocarbon and the like. Such a relatively inert gaseous substance may serve primarily as a temperature diluent which serves to obviate local overheating of the catalyst due to "hot spots", to obviate excessive oxidation of the catalyst and to minimize the occurrence of undesirable side reactions resulting in the formation of excessive amounts of carbon dioxide. In general, I prefer to execute my invention in the presence of steam or other relatively inert substances which normally exist in the liquid state, since such diluents are readily separated from the normally gaseous unreacted olefines by condensation.

My process comprises effecting the controlled catalytic oxidation of an olefine to the corresponding olefine oxide by causing said olefine to react with free oxygen, which oxygen may be present as oxygen per se or in admixture with other gaseous materials as steam, nitrogen, carbon dioxide, etc., containing, for example, from 21.0% to 99.9% oxygen, or the free oxygen may be liberated or formed under the conditions of operation from substances containing oxygen in combined form.

The present invention is, in the majority of cases, executed in the vapor phase. When vapor phase methods are employed, the gaseous olefine in admixture with the desired quantity of oxygen per se or oxygen contained in a mixture or solution with other gases as in air is passed preferably continuously over the stationary catalytic material at a predetermined space velocity. In general, one mol. of oxygen reacts with two mols of a mono-olefine to form two mols of the corresponding olefine oxide. I prefer, in the majority of cases, to conduct the reaction in the presence of an excess of olefine over the stoichiometrical amount needed to combine with all of the oxygen present in the reaction space. This excess of olefine may vary within wide limits depending on the activity of the catalyst employed and on the specific operating conditions. Excellent results may usually be obtained by operating so that the reactants enter the reaction chamber in the ratio of one mol. oxygen to about 2.0 to 2.5 mols of mono-olefine. When steam and the like diluents are employed the same may be introduced into the reaction chamber separately or in admixture with the reactants.

The term "space velocity", as used herein, may be defined as the unit volume of olefine flowing through the reaction space per unit volume of catalyst, under standard conditions of temperature and pressure.

My invention is executed in a preferred temperature range of from 200° to 500° C. The use of temperatures below about 200° C. is, in the majority of cases, impractical due to the relatively slow rate of reaction even when exceedingly low space velocities are employed. The practical upper limit of the reaction temperature is set by the occurrence of pyrolytic side reactions at too high a temperature at the existing pressure. The temperature to be chosen for each specific operation is dependent on the activity of the catalyst employed, on the pressure in the reaction chamber, on the space velocity, on the stability and character of the olefine reacted, and also on the stability of the resulting product as regards cracking, polymerization and resistance against further oxidation. When it is desired to employ high space velocities, it may be of advantage to operate at temperatures near the upper limit of the allowable range. If low space velocities are employed, it may be desirable to operate at temperatures near the lower limit of the range.

The reaction of an olefine with free oxygen to form an olefine oxide is in itself endothermic. However, due to the fact that the reaction to form the olefine oxide is usually accompanied by oxidation of the olefine and/or olefine oxide to carbon dioxide, heat will be liberated in the course of the reaction. The temperature of the catalytic mass may be kept substantially constant at the predetermined desired temperature by one or combinations of the suitable methods known to the art. By a proper method of distributing the gas mixture comprising the olefine, oxygen and possibly steam, nitrogen, etc., throughout the catalyst bed, the whole may, once the reaction is initiated, be kept at a uniform temperature without the application of external heat, thus effecting a considerable economy of operation. Furthermore, the space velocity may be adjusted and the heat of reaction thereby controlled, or suitable heat exchangers may be internally or externally applied. The reaction zone may be surrounded by a selected constant boiling substance which on vaporization consumes heat liberated by the reaction and thereby maintains a substantially constant temperature of the reaction zone. To constant boiling substance, if inert, may be directly introduced into the reaction zone, from which it may be recovered and reused. In the execution of my invention, I may employ the heat liberated by the oxidation reaction to preheat, severally or in combination, the gases entering the reaction zone, thus effecting thermal economies.

My process may be executed at subatmospheric, atmospheric or superatmospheric pressure. In general, I prefer to execute the same at pressures slightly in excess of atmospheric, although it can be seen that when operating with olefines which might themselves polymerize or yield readily polymerizable or decomposable reaction products at

the desired reaction temperature, it may be desirable to employ atmospheric or subatmospheric pressures.

The exit vapors from the reaction zone may be passed into a suitable condenser or fractionating column wherein the less volatile olefine oxide, water and other condensable constituents are separated from the unreacted olefine. After separation of the olefine oxide the reaction gases containing unreacted olefine may be mixed with the desired quantity of free oxygen or free oxygen containing gas and again passed over the catalyst to obtain further conversion of the olefine contained therein. The water formed in the reaction zone or introduced therein as steam, is generally liquefied with the olefine oxide, from which it may be separated by any suitable method as fractionation, stratification, use of drying agents, and the like. Any other normally liquid substance formed in the course of the reaction or introduced into the reaction mixture may be separated from the liquid olefine oxide by any suitable method known to the art.

For illustrative purposes only, reference is had to several examples which typify preferred methods of preparing suitable catalytic material and preferred modes and conditions of executing my invention.

Example I

An active finely divided silver catalyst was prepared by heating silver oxalate ($Ag_2C_2O_4$) at a temperature slightly above its decomposition temperature until substantially complete decomposition to metallic silver was effected.

An ethylene-air mixtufe containing from 8% to 9% ethylene was passed over the stationary silver catalytic material contained in a suitable reaction tube and heated to a temperature of about 350° C. The ethylene-air mixture was passed over the catalyst at the rate of about 88 liters per hour (measured at room temperature and atmospheric pressure) while steam was added at the rate of from 100 to 200 gm. per hour. During the run, the reaction temperature was maintained at from about 350° C. to about 400° C.

The exit vapors were condensed and the condensate containing water and ethylene oxide was fractionated for the recovery of the ethylene oxide.

About 10% of the ethylene was converted to ethylene oxide on one passage over the catalyst. The exit gases, after condensation of the ethylene oxide and water, were mixed with the desired quantity of air and again passed through the apparatus for further conversion of the ethylene.

This experiment was repeated using a colloidal silver catalyst and a catalytic silver prepared by reducing, with hydrogen, silver hydroxide precipitated on pumice stone or silica gel. With these catalysts, and employing substantially identical reaction conditions, only traces of ethylene oxide could be detected in the liquid condensate.

Example II

A very efficacious silver catalyst containing a small amount of gold which serves as a promoter was prepared as follows: the compound

$$[Au(NH_3)_4](NO_3)_3$$

was prepared by reacting ammonia and ammonium nitrate with gold chloride. 200 gm. of silver nitrate and 1.6 gm. of $[Au(NH_3)_4](NO_3)_3$ were dissolved in 2 liters of water and this solution was added to a solution of 84 gm. of sodium oxalate in 3 liters of water. The precipitate, which consisted of silver oxalate and the compound $[Au(NH_3)_4](C_2O_4)NO_3$, was successively washed with alcohol and ether and the ether evaporated therefrom by exposure to the air. The precipitate was then decomposed in small portions in a porcelain crucible heated to about 300° C.

A convenient quantity of the catalytic material was placed in a suitable reaction tube inclosed in a reaction furnace and maintained at a temperature of about 350° C. About 88 liters of a gas mixture having the approximate by volume composition: 0.2% CO_2, 7.8% C_2H_4, 19.3% O_2, 72.7% N_2 and about 174 gm. of steam were passed per hour over the catalyst.

About 17.4% of the ethylene present was converted to ethylene oxide on one passage over the catalytic material. Substantially complete conversion of the ethylene to oxide was effected by repeated admixture with oxygen and passage of the mixture over the catalyst.

Example III

About 100 gm. of silver nitrate and about 1.5 gm. of copper nitrate were dissolved in one liter of water and the resulting solution added to a solution of about 41 gm. of sodium oxalate in 1.5 liters of water. The precipitate was washed, dried, decomposed by heating and subsequently brought into a suitable reaction tube.

By using this catalyst and employing substantially the same reaction conditions as described in Example II, about 12% of the ethylene present in the reaction mixture was converted to ethylene oxide on one passage over the catalytic material.

Example IV

About 100 gm. of silver nitrate and about 0.215 gm. of ferric nitrate were dissolved in 1 liter of water and the resulting solution added to a solution of about 42 gm. of silver oxalate in about 1.5 liters of water. The resulting precipitate was separated from the aqueous solution, washed, dried and decomposed by heating.

The resulting catalyst was used under substantially the same conditions as described in Example II. An average conversion of from 12% to 13% of the ethylene to ethylene oxide was effected on one passage of the gas mixture through the reaction zone.

Example V

About 100 gm. of silver nitrate and about 5 gm. of manganese nitrate were dissolved in about 500 cc. of water and the solution was added to a solution of about 42 grams of sodium oxalate in about 1.5 liters of water. The resulting precipitate was washed successively with water, alcohol and ether, then freed of ether by exposure to the air and decomposed in small portions by heating. About 48 gm. of catalytic material having an apparent volume of about 220 cc. were obtained.

A gas mixture having the approximate by volume composition: 0.4% CO_2, 8.4% C_2H_4, 18% O_2 and 73.2% N_2 was treated as follows: the gas mixture was passed over the catalyst at an average rate of about 88 liters per hour, while steam was added at an average rate of about 200 gm. per hour. The catalytic material was maintained at an average temperature of about 350° C.

About 16.9% of the ethylene was converted

to ethylene oxide on one passage through the reaction zone.

My invention provides a batch, intermittent or continuous mode of operation whereby olefines may be substantially completely converted to olefine oxides.

It will be evident to those skilled in the art that the specific operating conditions such as space velocities, pressure, temperature, etc., may be varied within wide limits within the scope of my invention, depending on the relative activity of the catalyst employed, the concentrations of olefine, olefine oxide and free oxygen in the reaction mixture and on the specific olefine oxidized.

While I have in the foregoing described in some detail the preferred embodiments of my invention and some variants thereof, it will be understood that this is only for the purpose of making the invention more clear and that the invention is not to be regarded as limited to the details of operation herein described, nor is it dependent upon the soundness or accuracy of the theories which I have advanced as to the advantageous results obtained. On the other hand, the invention is to be regarded as limited only by the terms of the accompanying claims, in which it is our intension to claim all novelty inherent therein as broadly as possible in view of the prior art.

I claim as my invention:

1. A process for the production of olefine oxides which comprises reacting an olefine with oxygen in the presence of a silver catalyst obtained by effecting the thermal decomposition of silver oxalate, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

2. A process for the production of olefine oxides which comprises reacting an olefine with oxygen in the presence of an activated catalyst prepared by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid, and activated by the addition thereto of a metallic promoter essentially comprising a heavy metal, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

3. A process for the production of olefine oxides which comprises reacting an olefine with oxygen in the presence of an activated catalyst prepared by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid, and activated by the addition thereto of a heavy metal catalyst promoter of the class consisting of copper, gold, nickel, iron, cobalt, manganese, cerium, thorium and zinc, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

4. A process for the production of olefine oxides which comprises reacting an olefine with oxygen in the presence of an activated catalyst prepared by effecting the thermal decomposition of an intimate mixture of a silver salt of an aliphatic carboxylic acid with a small amount of a gold compound, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

5. A process for the production of olefine oxides which comprises reacting an olefine with less than the stoichiometrical amount of free oxygen in the presence of a catalyst essentially comprising silver metal obtained by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

6. A process for the production of olefine oxides which comprises reacting an olefine with oxygen in the presence of a relatively inert gas, said reaction being effected in the presence of a catalyst essentially comprising silver metal obtained by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

7. A process for the production of olefine oxides which comprises reacting an olefine with oxygen in the presence of steam, said reaction being effected in the presence of a catalyst essentially comprising silver metal obtained by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

8. A process for the production of olefine oxides which comprises reacting an olefine with oxygen at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored in the presence of a catalyst essentially comprising silver metal obtained by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid.

9. A process for the production of olefine oxides which comprises reacting an olefine with oxygen at temperatures between 200° C. and 500° C. in the presence of a catalyst essentially comprising silver metal obtained by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid.

10. A process for the production of olefine oxides which comprises reacting an olefine with oxygen in the molal ratio of at least two mols of olefine to one mol. of oxygen, in the presence of steam, at a temperature in the range of from 200° C. to 500° C. and in the presence of a catalyst essentially comprising silver metal obtained by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid.

11. A process for the production of olefine oxides by catalytically oxidizing olefines which comprises passing an olefine-air mixture at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored over a silver catalyst prepared by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid.

12. A process for the production of olefine oxides by catalytically oxidizing olefines which comprises passing an olefine-air mixture containing olefine and oxygen in the ratio of at least two mols of olefine to one mol. of oxygen, in the presence of steam and at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored, over a catalyst mass comprising silver metal obtained by effecting the ther-

mal decomposition of a silver salt of an aliphatic carboxylic acid.

13. A process for the production of ethylene oxide which comprises reacting ethylene with oxygen in the presence of a silver catalyst prepared by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

14. A process for the production of ethylene oxide which comprises reacting ethylene with oxygen in the presence of steam at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored and in the presence of a silver catalyst prepared by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid.

15. A process for the production of ethylene oxide which comprises reacting ethylene with oxygen in the presence of a silver catalyst obtained by effecting the thermal decomposition of silver oxalate, said reaction being effected at a temperature not substantially less than 200° C but below the temperature at which pyrolytic side reactions are substantially favored.

16. A process for the production of ethylene oxide which comprises reacting ethylene with oxygen in the presence of steam at a temperature in the range of from 200° C. to 500° C. and in the presence of a silver catalyst essentially comprising silver metal prepared by effecting the thermal decomposition of silver oxalate.

17. A process for the production of ethylene oxide by catalytically oxidizing ethylene which comprises passing an ethylene-air mixture containing ethylene and oxygen in the molal ratio of at least two mols of ethylene to one mol. of oxygen, in the presence of steam and at a temperature in the range of from 200° C. to 500° C., over a catalyst mass essentially comprising silver metal obtained by effecting the thermal decomposition of silver oxalate.

18. A process for the production of ethylene oxide which comprises reacting ethylene with oxygen in the presence of steam at a temperature in the range of from 200° C. to 500° C. and in the presence of an activated catalyst prepared by effecting the thermal decomposition of silver oxalate and activated by the addition thereto of a heavy metal catalyst promoter of the class consisting of copper, gold, nickel, iron, cobalt, manganese, cerium, thorium and zinc.

19. A process for the production of an olefine oxide which comprises reacting an olefine with oxygen in the presence of a silver metal catalyst obtained by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

20. A process for the production of an olefine oxide which comprises reacting an olefine with oxygen in the presence of a relatively inert diluent material and in the presence of a silver metal catalyst obtained by effecting the thermal decomposition of a silver salt of an aliphatic carboxylic acid, said reaction being effected at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored.

21. A process for the production of an olefine oxide which comprises contacting an olefine with less than a stoichiometrical amount of free oxygen and subjecting the mixture to a treatment, in the presence of a silver metal catalyst obtained by thermal decomposition of a silver salt of an aliphatic carboxylic acid, at a temperature of from about 200° C. to about 500° C.

22. A process for the production of ethylene oxide which comprises reacting ethylene with oxygen in the presence of steam at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored, said reaction being catalyzed by the presence of an active catalyst prepared by effecting the thermal decomposition of an intimate mixture of a silver salt of an aliphatic carboxylic acid with a relatively smaller amount of a gold compound.

23. A process for the production of ethylene oxide which comprises reacting ethylene with oxygen in the presence of steam at a temperature not substantially less than 200° C. but below the temperature at which pyrolytic side reactions are substantially favored, said reaction being catalyzed by the presence of an active catalyst prepared by effecting the thermal decomposition of an intimate mixture of a silver salt of an aliphatic carboxylic acid with a relatively smaller amount of a manganese compound.

ADRIANUS JOHANNES VAN PESKI.

PATENT SPECIFICATION

402,438

Convention Date (France): Oct. 3, 1931.

Application Date (In United Kingdom): May 2, 1932. No. 12,561/32.

Complete Accepted: Dec. 4, 1933.

COMPLETE SPECIFICATION.

Improvements in or relating to the Production of Ethylene Oxide, particularly for the Preparation of Ethylene-glycol and of its Derivatives.

We, SOCIETE FRANCAISE DE CATALYSE GENERALISEE, a Joint-stock company duly organised according to French Laws, of 8, Avenue-Percier, Paris, France, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

The invention relates to the preparation of ethylene oxide. It is known that ethylene may be converted into aldehydes, more particularly formaldehyde, by oxidation. The conversion of ethylene into ethylene oxide, however, has heretofore offered considerable difficulties. In fact, the separation of the aldehydes and the ethylene oxide constitutes a complicated operation which hitherto has not permitted the industrial preparation of ethylene oxide by the direct oxidation of ethylene. With the process according to the invention, ethylene oxide and carbon dioxide are obtained without aldehydes except in the form of traces.

The present invention comprises subjecting the ethylene to the simultaneous action of the oxygen of the air and water in the presence of a catalyst constituted by the following metals and their oxides, alone or in admixture; namely antimony, lead, bismuth, silver, arsenic, nickel, tin, gold, copper, iron.

It has been found that hydrogen, when introduced into the reaction, assists the latter.

Hydrogen probably plays the part of an agent for accelerating the base reaction of the process which can be written as follows:—

$$2\ \begin{matrix} H_2C \\ \| \\ H_2C \end{matrix} + O_2 \longrightarrow 2\ \begin{matrix} H_2C \\ \| \\ H_2C \end{matrix}\!\!>\!O$$

This reaction takes place at atmospheric pressure, but for commercially carrying the said process into practice, pressure can be employed, for instance a pressure of 50 kilograms per square centimeter, at a temperature between 150 and 400° C.

The following catalysts may be used:—

1.) a mixture of antimony, bismuth, silver, lead, arsenic, nickel and tin in variable proportions;

2.) silver alone,
bismuth alone,
antimony alone,

3.) mixtures of antimony, lead, nickel and silver,

4.) mixtures of bismuth, nickel, silver and lead,

5.) mixtures of antimony, bismuth, nickel,

6.) mixtures of antimony, bismuth, silver,

7.) mixtures of bismuth and nickel,

8.) mixtures of bismuth and silver.

And, generally speaking one or mixtures of the following metals: antimony, lead, bismuth, silver, nickel, tin, arsenic, gold, copper, iron, taken two by two, three by three, four by four, etc. . . in variable proportions, the mixtures, in the same conditions, of these metals with their oxides, the mixtures of the oxides alone of these metals, in the same conditions.

The ethylene can be obtained from any source of supply: gas from coke ovens, hydrogenation of acetylene, dehydration of ethyl alcohol, and the like.

Water can be admitted in the reaction vessel, either in the liquid state, or as steam.

The oxygen can be the oxygen of the air, this latter gas being introduced in the reaction.

Several modi operandi for carrying the process into practice, will be indicated hereinafter, by way of example only.

MODUS OPERANDI I.—

In a plant constructed for resisting high pressures, ethylene on the one hand, and air on the other hand, are separately compressed by means of suitable compressors.

The mixture of air and ethylene can also be compressed by taking the necessary precautions in order to avoid the risk of explosion.

These two gases will be led, together with hydrogen or not, into a reaction tube

[Price 1/-]

resisting the pressure of several hundreds of kilograms, containing one of the catalysers indicated above, and heated, for instance, to a temperature of 200° C. This tube communicates moreover with a circulating pump allowing to introduce water under pressure.

The reaction will take place in this tube between ethylene, the oxygen of air, and water. By controlling the outputs, a continuous operation will be obtained. Ethylene oxide thus formed will be separated from the other gases, for instance, by condensation.

Modus Operandi II.—

Silver in powder form, or in the form of a wire gauze, and placed or not on a support or carrier, is introduced in a tube, made for instance of silica, and, after the said tube has been heated to a temperature comprised between 150 and 400° C., a mixture of ethylene, air, water vapour, and hydrogen is caused to pass therethrough. The proportion of hydrogen is about 5% of the total mixture. The ethylene oxide produced can be separated from the gases issuing from the tube.

Modus Operandi III.—

Colloidal silver is introduced in a high pressure tube which has been partly filled with water. Pure ethylene under pressure is then added in order that it can dissolve in the water.

The tube is subsequently heated between 150° and 400° C., then air and hydrogen are slowly introduced. The introduction of air is stopped when the totality of ethylene is converted into ethylene oxide.

From ethylene oxide produced according to the processes which have been indicated above, it is possible to obtain by simple saponification, ethylene-glycol, or by other known methods any derivative of ethylene-glycol.

The reaction, for obtaining ethylene-glycol can be written as follows:

$$C_2H_4O + H_2O \longrightarrow \begin{matrix} H_2C{-}OH \\ | \\ H_2C{-}OH \end{matrix}$$

The experiments effected by the applicant have shown that, in presence of the catalysers indicated, water, in the form of steam or not, considerably promotes the reaction ensuring the production of ethylene oxide.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1. A process of obtaining ethylene oxide consisting in subjecting ethylene to the simultaneous action of the oxygen of the air and water, in the presence of a catalyst constituted by the following metals and their oxides, alone or in admixture: namely antimony, lead, bismuth, silver, arsenic, nickel, tin, gold, copper, iron.

2. A process for the production of ethylene oxide, which consists in subjecting ethylene to the simultaneous action of oxygen and water, in the presence of a catalyst, constituted by a mixture of antimony, bismuth, silver, lead, arsenic, nickel and tin, at a temperature between 150 and 400° C.

3. A process for the production of ethylene oxide as claimed in claims 1 and 2, characterised by the fact that a mixture of ethylene, air, water vapour and hydrogen is caused to pass in a tube, made of refractory material, containing the catalyst and heated to a temperature comprised between 150 and 400° C.

4. A process for the production of ethylene oxide as claimed in claim 3, characterised by the fact that the proportion of hydrogen is about 5% of the total volume of the gases in presence.

5. The process for the production of ethylene oxide, particularly for the preparation of ethylene-glycol and of its derivatives, substantially as hereinbefore described.

Dated this 2nd day of May, 1932.

SOCIETÉ FRANCAISE DE CATALYSE GENERALISEE,
Per Boult, Wade & Tennant,
111 & 112, Hatton Garden, London, E.C. 1,
Chartered Patent Agents.

Redhill: Printed for His Majesty's Stationery Office, by Love & Malcomson, Ltd.—1933.

Defendant's Exhibit 73

PATENT SPECIFICATION

Convention Date (France): Dec. 19, 1931.

402,749

Application Date (In United Kingdom): May 2, 1932. No. 12,579/32.

(Patent of Addition to No. 402,438. Convention Date (France): Oct. 3, 1931.)

Complete Accepted: Dec. 4, 1933.

COMPLETE SPECIFICATION.

Improvements in or relating to the Production of Ethylene Oxide, particularly for the Preparation of Ethylene-glycol and of its Derivatives.

We, SOCIETE FRANCAISE DE CATALYSE GENERALISEE, a Joint-Stock Company duly organised according to French Laws, of 8, Avenue Percier, Paris, France, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

The process described in the main British patent for "Process for the production of ethylene oxide, particularly for the preparation of ethylene-glycol and of its derivatives", dated 2nd May, 1932, No. 12,561/32 (Serial No. 402,438) mainly consists in subjecting ethylene to the simultaneous action of oxygen and water in presence of a catalyst and if desired of hydrogen.

In the examples described in the main patent for carrying this process into practice, the reaction takes place at a pressure equal to or greater than atmospheric pressure, in presence of catalysts among which is cited silver in the colloidal form, or in the form of a powder or of a gauze.

Experiments have shown that silver could be considerably activated by introducing, into this body, small quantities of gold, copper and iron.

According to the invention, therefore, a process for the production of ethylene oxide consists in subjecting ethylene to the simultaneous action of oxygen and water in the presence of a catalyst constituted by silver containing small quantities either of gold, or of copper, or of iron, at a temperature between 150 and 400° C.

Thus, a mixture containing silver, gold, copper and iron, gives very satisfactory results when used as a catalyst and allows a specific reaction; the only foreign product obtained in the reaction is CO_2, which can be easily removed. Aldehydes formed exist only as traces.

The applicant has also found that the reaction giving CO_2 is, contrary to previous belief, a reaction of oxidation independent from that giving ethylene oxide and from that giving aldehydes. From experiments effected by the applicant, it results that, if water is introduced in suitable quantity, the reaction is not only facilitated, as indicated in the main Patent, but, in addition, the reaction giving CO_2, probably by direct oxidation of ethylene according to the equation:

$$C_2H_4 + 3O_2 + 2H_2O + 2CO_2$$

is checked, owing, as is probable, to the partial pressure of water. From these experiments, it can be concluded that if a certain quantity of CO_2 is previously introduced, in addition, in the reacting gases, the partial pressure of CO_2 will produce its effect and the reaction of oxidation of ethylene into CO_2 will be completely checked resulting in a considerable increase in the efficiency of the operation. Moreover, it is to be noted that, when the synthetic operation is effected under partial vacuum, the yield in ethylene oxide is increased.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1. A process for the production of ethylene oxide, consisting in subjecting ethylene to the simultaneous action of oxygen and water in the presence of a catalyst constituted by silver containing small quantities either of gold, or of copper, or of iron, at a temperature between 150 and 400° C.

2. A process for the production of ethylene oxide as claimed in Claim 1, in which there is added to the gases present, hydrogen in the proportion of 5% of the total volume of the said gases.

3. A process for the production of ethylene oxide as claimed in Claims 1 and 2, in which carbon dioxide gas is added to the gases present.

84. [Price 1/-]

Defendant's Exhibit 73 (continued)

4. A process for the production of ethylene oxide as claimed in Claim 1 and either of Claims 2 and 3, in which the reaction takes place at a pressure lower than atmospheric pressure.

Dated this 2nd day of May, 1932.

SOCIETE FRANCAISE DE CATALYSE GENERALISEE,
Per Boult, Wade & Tennant,
111 & 112, Hatton Garden, London, E.C. 1,
Chartered Patent Agents.

Redhill: Printed for His Majesty's Stationery Office, by Love & Malcomson, Ltd.—1933.

PATENT SPECIFICATION

Convention Date (France): July 10, 1933. 431,966

Application Date (In United Kingdom): July 9, 1934. No. 20089/34.

Complete Specification Accepted: July 18, 1935.

COMPLETE SPECIFICATION

Improvements in or relating to the Manufacture of Ethylene Oxide

We, SOCIETE FRANCAISE DE CATALYSE GENERALISEE, a Joint Stock Company duly organised under the French Laws, of 26 rue Pierre-Lhomme, Courbevoie (Seine) France, Manufacturers, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

This invention relates to the production of ethylene oxide by the catalytic oxidation of ethylene with oxygen or an oxygen-containing gas such as air.

This direct oxidation of ethylene requires special precautions owing to the danger of explosion. It has already been proposed to introduce water into the reacting mixture when carrying out the reaction on an industrial scale. Although this avoided the danger of explosions the products of the reaction were aldehydes and not ethylene oxide.

Another proposal has been to effect the catalytic oxidation of ethylene by treating ethylene with oxygen of the air and water and, in some cases, to add hydrogen to the reacting mixture. Since air was to be used there would be an appreciable amount of inactive gas, namely nitrogen, present in the reacting mixture.

A further proposal has been to effect the catalytic oxidation of ethylene by means of oxygen and water in the presence of carbon dioxide.

The advantageous action of carbon dioxide was stated to be explained by the following consideration:—

In addition to the oxidation yielding ethylene oxide, carbon dioxide is formed to some extent according to the equation

$$C_2H_4 + 3O_2 = 2CO_2 + 2H_2O$$

The reaction in this direction would appear to be suppressed or limited by the partial pressure of the carbon dioxide present in the reacting mixture and consequently the yield of ethylene oxide produced according to the equation

$$2C_2H_4 + O_2 = 2C_2H_4O$$

is increased by adding carbon dioxide to the reacting mixture.

The present invention is based on the discovery that an inert gas can be used to replace water as a diluent in the catalytic oxidation of ethylene and is therefore differentiated from the previous proposals by the use of a substantially dry reacting mixture.

A proposal has previously been made to oxidise olefines directly with air but only in vessels having a large volume ratio to ensure a homogeneous gas reaction as distinct from a heterogeneous or catalytic reaction.

Thus, according to the present invention, ethylene oxide is produced by the catalytic oxidation of ethylene in the presence of an inert gas and in the absence of water.

The addition of an inert gas to the reacting mixture as the only diluent has been found to have the same action as water in preventing explosion and has the further advantage that the reaction can be effected under increased pressure. When using water vapour and increased pressures up to fifty atmospheres it is impossible to maintain the water in the state of vapour unless undesirable high temperatures are employed.

Carbon dioxide is preferably used as the inert diluent and this has the additional advantage of inhibiting the undesirable oxidation of ethylene to carbon dioxide and water as indicated above.

A further advantage of substituting an inert gas for water is that the lower specific heat of such gas results in a considerable saving of heat in carrying out the reaction.

The following examples illustrate the manner in which the invention may be carried into effect.

EXAMPLE 1.

A gaseous mixture is prepared containing 66% carbon dioxide, 3, 4% ethylene and 30, 3% air and is passed by any suitable means e.g. by pumping from a gasholder to a catalyst chamber heated to a temperature between 300 and 400°C. Catalyst chamber is packed with a catalyst which may be pure silver or silver activated by the presence of small amounts of gold or copper or of both metals.

We have found that these catalysts namely silver alone, silver and gold, silver and copper or silver, gold and copper together are the most suitable for the production of ethylene oxide. After

passing over the catalyst the gases are treated to recover the ethylene oxide which has been produced. For example the oxide of ethylene may be absorbed by active carbon, or dissolved in water under normal or increased pressure or by any other known means. The carbon dioxide may be recovered by liquefaction and in this manner easily separated from the nitrogen. By a suitable regulation of the length of the catalyst layer and of the speed of passage of the reacting gases, the whole of the ethylene present and the whole of the oxygen can be caused to react. After liquefaction of the carbon dioxide the residual nitrogen can be used in any convenient manner, e.g. for the synthesis of ammonia. With a catalyst tube 23 millemeters in diameter and 60 centimeters length packed with catalyst we have found that the speed of passage of the gaseous mixture may vary between 85 and 250 litres per hour.

EXAMPLE 2.

The same gaseous mixture as described in example 1 is sent by means of a compressor to a steel tube capable of withstanding high pressure and heated electrically. Pressures up to 50 atmospheres may be employed. The heated tube is charged with one of the catalysts indicated hereinbefore. Temperature within the limits indicated in example 1 may be varied according to the pressure employed. The rate of gas flow may lie between 85 and 300 litres per hour.

The use of increased pressure favours the reaction giving rise to ethylene oxide since this reaction is effected with a resulting decrease in volume as can be seen from the equation

$$2\,C_2H_4 + O_2 = 2\,C_2H_4O$$

on the other hand the reaction giving rise to carbon dioxide and water is independent of the pressure, under those conditions of temperature and pressure in which the water is in the form of vapour. The presence of a large excess of carbon dioxide in the reacting gases becomes of greater importance when the synthesis is effected under pressure owing to the partial pressure of this carbon dioxide. The treatment of the products of reaction may be effected as described in the first example given.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1. The method of producing ethylene oxide by the catalytic oxidation of ethylene in the presence of an inert gas and in the absence of water.

2. Method according to claim 1 in which the inert gas is carbon dioxide.

3. Method according to claim 1 in which the catalyst consists of or comprises silver with the addition of gold and/or copper.

4. Method according to claim 1 in which the oxidation is carried out under increased pressure.

5. Method according to claim 1 in which the oxidation is effected at temperatures between 300 and 400°C.

6. Method according to claim 1 in which the proportion of inert gas is at least 50% of the total amount of the reacting gases.

7. Ethylene oxide whenever produced by the process of any of the preceding claims or by its obvious chemical equivalent.

Dated this 9th day of July, 1934.

For Societe Francaise de Catalyse Generalisee,
STEVENS, LANGNER, PARRY & ROLLINSON,
Chartered Patent Agents,
5-9, Quality Court, Chancery Lane,
London, W.C.2, and at
17, John Street, New York, U.S.A.

Leamington Spa: Printed for His Majesty's Stationery Office, by the Courier Press.—1935.

DEFENDANT'S EXHIBIT 76

FRENCH REPUBLIC
MINISTRY OF COMMERCE AND INDUSTRY
DEPARTMENT OF INDUSTRIAL PROPERTY

Patent of Invention

Group 14— Class 1 No: 771,650

PROCESS FOR OBTAINING ETHYLENE OXIDE BY THE DIRECT OXIDATION OF ETHYLENE.

Societe Anonyme:

SOCIETE FRANCAISE DE CATALYSE GENERALISEE

Residing in France (Seine)

Filed July 10, 1933, at 4:36 p.m. in Paris

Granted July 30, 1934—Published Oct. 13, 1934.

(Patent of invention the grant of which has been deferred in accordance with article 11, paragraph 7 of the law of July 5, 1844, as amended by the law of April 7, 1902.)

The present invention relates to the preparation of ethylene oxide by the direct oxidation of ethylene in contact with pure oxygen or air.

The reaction, or direct oxidation, requires special precautions against explosion hazards. In connection with the industrial oxidation of ethylene it was suggested, as early as 1904, that water vapor be introduced into the reaction. The explosion hazards are thus caused to disappear but the oxidation products have always been aldehydes.

The applicant has improved this process by bringing into play catalysts suitable for obtaining a specific reaction which gives ethylene oxide as the oxidation product. The thus improved process marks a considerable step forward in the technique of the production of ethylene oxide by the direct oxidation of ethylene, but the necessity

of vaporizing large amounts of water requires an expenditure of calories which obviously contributes its share to the production cost of the ethylene oxide obtained.

The present invention remedies this drawback. It has as its object a process for preparing ethylene oxide by the direct oxidation of ethylene without water vapor and with no explosion hazards.

This process is characterized principally in that the ethylene-oxygen or ethylene-air mixture is diluted with a large amount of an inert gas.

In one particularly advantageous form of application of the process defined above as to its general characteristics, the inert gas used is carbon dioxide.

The utilization of CO_2 as an inert gas leads in fact to an important industrial result which it seems should be explained as follows:

In addition to the oxidation of the ethylene, CO_2 is formed which can be considered as the ultimate limit of the oxidation reaction. This total oxidation is graphically represented as follows:

$$C_2H_4 + 3O_2 = 2CO_2 + 2H_2O$$

This reaction is reduced or limited by the partial pressure of the CO_2 present in a large proportion in the active mixture. The yield of the oxidation reaction giving the ethylene oxide is thus improved.

Moreover, the step of introducing CO_2 initially into the gaseous mixture has the considerable advantage of rendering said mixture non-explosive under the conditions in which the synthesis is brought about.

Finally, the substitution of CO_2 for water in the form of vapor enables one readily to effect the synthesis of ethylene oxide under pressure. With water, it is very difficult to superheat at the reaction temperature so as to have water present in the form of vapor notwithstanding the pressure used (from 0 to 50 kilograms per sq. cm.).

By way of example merely there will be described below two methods of operation for carrying the invention into practice:

First method of operation (under atmospheric pressure).

A mixture is made up containing for example 66% CO_2, 3.4% ethylene, 6.3% oxygen, and 24% nitrogen (mixture air + ethylene + CO_2).

This mixture, stored in a gas tank, is passed through the medium of a pump into a synthesis tube heated electrically for example and containing a catalyst. The reaction temperature is between 300 and 400° C. The catalyst used is pure silver, or silver activated either by small amounts of gold or copper or by both gold and copper.

The four catalysts which at present appear to be the most appropriate ones, are:

1) Pure Ag
2) Ag + Au
3) Ag + Cu
4) Ag + Au + Cu.

After their passage through the synthesis tube containing one of the aforementioned catalysts, the gases are passed into a device which enables one to recover the ethylene oxide formed.

These means may either be activated carbon which absorbs the ethylene oxide, or water, whether under pressure or not, which dissolves the ethylene oxide formed, or any other means. The CO_2 which has served can be readily recovered, as for example by liquefaction, whereby it can be readily separated from the nitrogen. In fact, by suitably regulating the length of the catalyst contained in the synthesis tube, and the velocity of the gaseous reactants, all the ethylene and all the oxygen can be made to disappear. After the liquefaction of the CO_2 there thus remains pure nitrogen which can be used, for example in the synthesis of ammonia.

In tests conducted by the applicant the velocity of the gaseous mixture varied between 85 and 250 liters per

hour for a catalytic distance of 60 cm. The synthesis tube was 23 mm. in diameter, and the reaction temperature was about 375° C.

Second method of operation. (Under pressure). One makes up, for example, the same mixture as is described in the first method of operation. This mixture is then delivered by means of a compressor into a steel tube capable of resisting the pressure and heated, for example, electrically. The pressure used may vary between 0 and 50 kg. per sq. cm.

The heated steel tube contains a catalyst selected from among the four specified above.

The temperature used varies within the limits specified above, these variations corresponding to the pressure used and to the rate of gas flow which ranges between 85 and 300 liters per hour.

It is to be noted that pressure promotes the reaction yielding the ethylene oxide. In fact, it will be seen that this reaction

$$2C_2H_4 + O_2 = 2C_2H_4O$$

takes place with contraction of volume, whereas the total oxidation reaction yielding CO_2 and water according to

$$C_2H_4 + 3O_2 = 2CO_2 + 2H_2O$$

is independent of the pressure under the pressure conditions under which the water is in the form of a vapor at the reaction temperature. Moreover, the large excess of CO_2 will assume an even greater importance in the case of the pressure owing to its partial pressure.

The ethylene oxide formed is recovered, as stated above, either by water in order to dissolve it, or by activated carbon in order to absorb it, or by any other means.

One can likewise recover the CO_2 and at the end of the operation obtain pure nitrogen utilizable for all synthesis.

Resumé

The invention has for its object a process for obtaining ethylene oxide by the direct oxidation of ethylene by

means of pure oxygen or air, characterized principally in that the ethylene-oxygen or ethylene-air mixture is diluted with a large proportion of an inert gas.

Methods of carrying the invention into practice are characterized in that:

a) The inert gas used is nitrogen;

b) The inert gas is carbon dioxide which owing to its partial pressure limits the harmful reaction yielding CO_2 and water.

c) The oxidation reaction takes place in the presence of a catalyst constituted by pure silver, silver activated by small amounts of gold or copper, or both gold and copper.

d) The reaction takes place under atmospheric pressure;

e) The reaction takes place under a pressure which may reach 50 kg. per sq. cm.

Societe Anonyme: SOCIETE FRANCAISE DE CATALYSE GENERALISEE

By Power of Attorney
ELLUIN AND BARNAY.

DEFENDANT'S EXHIBIT 78

Liber B168 page 547

ASSIGNMENT

WHEREAS, SOCIETE FRANCAISE DE CATALYSE GENERALISEE, a French limited company having an office at No. 26 rue Pierre Lhomme, Courbevoie, France, is the sole and exclusive owner of Letters Patent of the United States No. 1,998,878 issued April 23, 1935, and entitled "Processes for the Production of Ethylene Oxide", and Application for Letters Patent

of the United States No. 39,196, entitled "Improvements relative to the Production of Olefine Oxide and of Catalysts for Use in such Production"; and

WHEREAS, CARBIDE AND CARBON CHEMICALS CORPORATION, a corporation organized and existing under the laws of the State of New York, having an office at No. 30 East 42nd Street, in the City and State of New York, United States of America, is desirous of acquiring the entire right, title and interest in and to and under the said Letters Patent and Application for Letters Patent and the inventions covered thereby, and any and all improvements at any time made thereon:

NOW, THEREFORE, TO WHOM IT MAY CONCERN, be it known that for and in consideration of the sum of One Dollar ($1.00) to it in hand paid by said CARBIDE AND CARBON CHEMICALS CORPORATION, and other good and valuable considerations, the receipt of which is hereby acknowledged, said SOCIETE FRANCAISE DE CATALYSE GENERALISEE has sold, assigned, transferred and set over and does hereby sell, assign, transfer and set over to said CARBIDE AND CARBON CHEMICALS CORPORATION, its successors, assigns and other legal representatives, the entire right, title and interest in and to said Letters Patent and said Application for Letters Patent and the inventions therein disclosed, described or claimed, and in and to any and all improvements at any time made thereon, and in and to any and all Letters Patent of the United States which may be granted (if any) thereon, and all reissue or reissues, division or divisions, or extension or extensions of any such Letters Patent, the same to be held and enjoyed by the said CARBIDE AND CARBON CHEMICALS CORPORATION for its own use and behoof and for the use and behoof of its successors, assigns or other legal representatives, to the end of the term or terms for which said Letters Patent may be granted, reissued, divided or extended, as fully and entirely as the same would have been held and enjoyed by said SOCIETE FRANCAISE DE CATALYSE GENERALISEE if this assignment and sale had not been made, together with all claims for damages by reason of past infringement of said Letters Patent with

the right to sue for and collect the same for its own use and behoof and for the use and behoof of its successors, assigns or other legal representatives.

And said SOCIETE FRANCAISE DE CATALYSE GENERALISEE hereby authorizes and requests the Commissioner of Patents to issue any and all Letters Patent of the United States under said invention or improvements or resulting from said Application or any divisions thereof to said CARBIDE AND CARBON CHEMICALS CORPORATION as Assignee of the entire interest.

And said SOCIETE FRANCAISE DE CATALYSE GENERALISEE further covenants and agrees that it will at any time upon request execute and deliver any and all papers that may be necessary or desirable to perfect the title to said invention or improvements thereon, or any Letters Patent that may be granted therefor, in CARBIDE AND CARBON CHEMICALS CORPORATION, its successors, assigns or other legal representatives, and that if the said CARBIDE AND CARBON CHEMICALS CORPORATION, its successors, assigns or other legal representatives, desires to secure a reissue, division or extension of said Letters Patent it will, upon request, sign all papers, make all rightful oaths and do all legal acts necessary and requisite for the application of such reissue, division or extension and the procuring thereof, but at the expense of CARBIDE AND CARBON CHEMICALS CORPORATION, its successors, assigns or other legal representatives, and said SOCIETE FRANCAISE DE CATALYSE GENERALISEE hereby covenants that it has full right to convey the entire interest herein assigned and covenants and agrees that it will at any time, upon request, communicate to the said CARBIDE AND CARBON CHEMICALS CORPORATION, its successors, assigns or other legal representatives, any facts relating to the said invention or improvements thereon and Letters Patent or to the history thereof known to it, and will cause testimony to be given as to the same in any interference or litigation when requested so to do.

IN WITNESS WHEREOF SOCIETE FRANCAISE DE CATALYSE GENERALISEE has caused these presents to be executed by its Administrator thereunto

duly authorized and its Corporate Seal to be hereto affixed and attested this 18th day of August 1936.

SOCIETE FRANCAISE DE
CATALYSE GENERALISEE

By JACQUES FRANCON
Administrateur Directeur

(Rubber stamp)

Societe Francaise
De Catalyse Generalisee
26 Rue Pierre-Lhomme
Courbevoie (Seine)

Attest:

LUCIENNE BOUVERET

REPUBLIC OF FRANCE
CITY OF LYON
CONSULATE OF THE UNITED
STATES OF AMERICA } ss:

On this 18th day of August 1936, before me personally appeared Jacques Francon to me personally known and known to me to be the Administrator of Societe Francaise De Catalyse Generalisee, the company which executed the foregoing Assignment, and being by me duly sworn did acknowledge to me that he executed the same on behalf of Societe Francaise De Catalyse Generalisee for the purpose therein set forth, being thereunto duly authorized.

Aug 18, 1936
Stamp

GEO. P. WILSON

Vice Consul of the United States of America
LYON FRANCE

RECORDED
Transfers of Patents
U.S. Patent Office
Sept. 16, 1936
Liber B168, page 547
Conway P. Coe, Commissioner of Patents

Defendant's Exhibit 80

have similar effects in the polymorphic and stability relations of compounds with a common anion.

7. Binary crystals increase in hardness as the ionic potentials of their constituents increase.

BALTIMORE, MARYLAND

[CONTRIBUTION FROM THE SCHOOL OF CHEMISTRY OF THE UNIVERSITY OF MINNESOTA]

THE CATALYTIC ACTIVITY OF METALLIZED SILICA GELS. V. THE OXIDATION OF ETHYLENE[1]

BY L. H. REYERSON AND L. E. SWEARINGEN

RECEIVED MAY 31, 1928 PUBLISHED NOVEMBER 6, 1928

Previous investigations of the authors[2] have shown that metallized silica gels exhibit considerable activity in certain oxidation reactions. The following investigation is an extension of this work to the oxidation of ethylene. Willstätter and Bommer[3] in their quantitative study of the conditions for the formation of formaldehyde from ethylene showed that, unless dilute, both ethylene and formaldehyde are unstable at temperatures much above 300°. They found that the use of catalysts accelerated the oxidation of ethylene but no formaldehyde was detected when catalysts were used. An osmium catalyst was found to initiate the oxidation of ethylene at about 130°, while copper proved to be effective at 250°. Blair and Wheeler[4] continued these investigations and found that, with a platinum catalyst, formaldehyde was formed at 405° when the gases were streamed through the catalyst very rapidly. As a result of these investigations, it was hoped that the use of metallized silica gels as catalysts would so lower the temperature at which reaction occurred that partial oxidation of ethylene could be effected and a mechanism for its catalytic oxidation obtained.

The experimental procedure was similar to that used by the authors in the study of the oxidation of methane.[5] Samples of the same catalysts were used as in the preceding investigations. Mixtures of ethylene, oxygen and nitrogen were passed over the catalysts at temperatures ranging from about 100 to 310° at varying rates of flow. In no case did the sum of oxygen and ethylene in the mixture exceed 50% of the gas volume. Preliminary experiments showed that the products of oxidation were carbon dioxide and water, so that the gases after passing through the catalyst were analyzed for ethylene, oxygen and carbon dioxide. In no case did tests show the presence of intermediate products of oxidation.

[1] In memory of Ira Remsen.

[2] Swearingen and Reyerson, *J. Phys. Chem.*, 32, 113, 192 (1928).

[3] Willstätter and Bommer, *Ann.*, 422, 36 (1921).

[4] Blair and Wheeler, *J. Soc. Chem. Ind.*, 41, 303T (1922).

[5] *Ref.* 2, p. 192.

Defendant's Exhibit 80 (continued)

Experimental Results

The results of the experiments are given in Tables I to IV and Figs. 1 and 2 graphically represent typical experiments. The symbols used in the tables have the following significance.

A_0 = the percentage of ethylene in the original mixture
A' = the percentage of ethylene found in the exit gas
A'' = the percentage of ethylene calculated to be present
O_2 = the amount of oxygen present in the original mixture
O_2' = the amount of oxygen found in the exit gas
O_2'' = the amount of oxygen calculated to be present
f = the fraction of carbon dioxide in the final mixture
F = the percentage of carbon dioxide in the final mixtures ($F = 100f$)
x = the fraction of the ethylene oxidized
X = the percentage of ethylene oxidized ($X = 100x$)
B = temperature of bath surrounding catalyst tube
C = temperature of catalyst during reaction

TABLE I

SILVER CATALYST

Gas mixture: (A) Ethylene, 27.00%; (O_2) Oxygen, 18.00%

Temperature B	C	Rate, cc. per min.	Final gas analysis A'	O_2'	F	Calcd. X	A''	O_2''
310	313	30	25.90	12.89	3.79	6.63	26.13	12.99
310	311	50	25.85	14.82	2.87	5.17	26.34	14.21
310	312	60	26.25	15.05	2.51	4.55	26.42	14.68
310	313	180	26.10	17.86	1.32	2.41	26.70	16.32
200	201	30	26.50	18.20	0.66	1.22	26.85	17.13
200	201	60	26.42	18.28	.66	1.22	26.85	17.13
200	200	180	26.85	18.62	.44	0.87	26.80	17.42
	Gas mixture:		(A) Ethylene, 19.00%; (O_2) Oxygen, 26.50%					
300	302	30	17.30	19.25	4.24	10.72	17.60	20.76
300	301	60	18.20	21.06	3.56	9.01	17.90	22.12
300	301	100	18.41	23.51	2.33	6.00	18.28	23.62
300	302	140	18.73	24.30	1.94	5.00	18.40	24.10
300	304	180	18.96	22.48	1.52	3.94	18.53	24.64

TABLE II

COPPER CATALYST

Gas mixture: (A) Ethylene, 30.00%; (O_2) Oxygen, 18.00%

Temperature B	C	Rate, cc. per min.	Final gas analysis A'	O_2'	F	Calcd. X	A''	O_2''
310	310	30	29.80	1.87	11.02	16.65	27.80	3.48
310	312	60	28.80	6.16	8.80	13.50	23.24	6.40
310	324	100	29.50	8.80	6.64	10.37	28.68	9.25
310	327	180	29.45	12.55	4.26	6.84	29.15	12.37
	Gas mixture:		(A) Ethylene, 24.00%; (O_2) Oxygen, 19.20%					
290	290	30	23.50	16.38	2.09	4.26	23.46	16.47
290	290	95	24.20	18.04	0.98	2.03	23.75	17.92
290	290	180	24.61	17.08	0.76	1.57	23.80	18.21

Defendant's Exhibit 80 (continued)

TABLE II (*Concluded*)

Gas mixture; (A) Ethylene, 19.50%; (O_2) Oxygen, 27.50%

Temperature B	Temperature C	Rate, cc. per min.	Final gas analysis A'	Final gas analysis O_2'	Final gas analysis F	Calcd. X	Calcd. A''	Calcd. O_2''
300	306	30	14.45	5.58	17.76	38.70	14.09	5.75
300	317	60	15.72	6.50	14.42	32.30	15.10	9.82
300	330	140	16.88	13.68	10.62	24.65	16.26	14.48
300	320	180	17.25	16.40	8.84	20.85	16.80	16.65
200	200	30	19.00	24.65	2.13	5.36	18.85	24.89
200	200	60	19.23	25.02	1.85	4.64	18.94	25.55
200	205	180	19.43	26.10	0.96	2.45	19.20	26.32
300		30	27.80	3.87	11.02	16.65	27.80	3.48
260		30	29.55	12.25	4.53	7.19	29.10	12.08
200		30	23.56	16.39	2.09	4.26	23.46	16.47
150		30	29.20	20.18	1.18	2.00	28.75	19.78
106		30	23.75	18.73	0.54	1.14	23.86	18.90

TABLE III

PLATINUM CATALYST

Gas mixture: (A) Ethylene, 20.00%; (O_2) Oxygen, 18.00%

Temperature B	Temperature C	Rate, cc. per min.	Final gas analysis A'	Final gas analysis O_2'	Final gas analysis F	Calcd. X	Calcd. A''	Calcd. O_2''
210	227	60	17.08	7.93	7.62	17.65	17.72	7.97
203	212	60	17.78	9.00	7.02	16.38	17.90	8.76
198	212	60	18.00	10.37	6.13	14.45	18.16	9.90
170	175	60	18.60	13.14	3.94	9.45	18.82	12.80
160	165	60	18.85	14.72	2.60	6.33	19.22	14.56
130	125	60	19.50	16.78	1.31	3.23	19.61	16.27
115	115	60	19.95	17.30	0.64	1.59	19.81	17.15
90	90	60	19.85	18.07	0.22	0.55	19.95	17.71

Gas mixture: (A) Ethylene, 29.00%; (O_2) Oxygen, 21.20%

200	215	30	26.45	4.40	12.53	19.22	26.48	5.18
200	215	60	27.30	8.80	10.10	15.90	27.88	8.30
200	220	100	27.06	10.63	7.98	13.00	27.32	11.02
200	225	140	27.50	13.09	6.15	10.08	27.71	13.38
200	235	180	27.99	14.10	5.50	8.97	27.85	14.22
130	130	30	28.43	18.59	2.18	3.67	28.44	18.49
130	130	60	28.68	20.42	0.76	1.30	28.84	20.32
130	130	180	28.70	20.65	0.54	0.93	28.89	20.51

Gas mixture: (A) Ethylene, 21.50%; (O_2) Oxygen, 26.50%

300	300	30	15.50	1.18	21.00	40.40	15.50	0.55
300	308	60	16.00	2.24	20.08	38.00	15.80	1.75
300	329	120	16.40	5.18	17.06	33.95	11.64	5.45
300	335	180	17.78	7.35	15.35	31.00	17.31	7.55
200	200	30	15.53	4.23	18.35	36.10	16.26	3.80
200	217	60	16.50	5.03	18.06	35.60	16.36	4.25
200	223	180	17.75	11.41	12.80	26.40	17.85	10.68
110	110	30	21.00	25.80	0.76	1.75	21.29	25.50
95	95	30	21.78	25.90	0.66	1.64	21.31	25.68
90	90	30	21.00	25.95	0.55	1.36	21.34	25.82

Defendant's Exhibit 80 (continued)

TABLE III (*Concluded*)

Gas mixture: (A) Ethylene, 21.30%; (O_2) Oxygen, 16.85%

Temperature B	Temperature C	Rate, cc. per min.	Final gas analysis A'	Final gas analysis O_2'	Final gas analysis P	Calcd. X	Calcd. A''	Calcd. O_2''
300	304	60	17.68	1.34	12.13	26.45	17.93	0.75
300	306	60	17.96	1.54	12.00	25.15	17.84	0.87
250	255	60	18.63	6.02	8.33	18.00	18.91	5.75
200	203	60	19.95	10.85	5.76	12.77	19.65	9.19
150	150	60	20.39	12.36	3.57	8.10	20.28	12.08
100	100	60	20.56	14.29	1.75	4.02	20.80	14.52
Gas mixture: (A) Ethylene, 22.65%; (O_2) Oxygen, 17.50%								
310	315	60	19.20	2.07	11.65	23.08	19.46	2.07
280	285	60	19.00	4.83	10.60	20.10	19.92	4.25
260	262	60	20.25	7.67	7.80	16.00	20.51	7.17
220	222	60	20.85	11.13	5.18	10.85	21.83	10.64
134	135	60	22.50	15.70	1.53	3.33	22.23	15.47
120	120	60	22.50	16.64	1.08	2.36	22.35	16.07
95	95	60	22.62	16.73	0.86	1.88	22.41	16.36

TABLE IV

PALLADIUM CATALYST

Gas mixture: (A) Ethylene, 24.00%; (O_2) Oxygen, 20.50%

Temperature B	Temperature C	Rate, cc. per min.	Final gas analysis A'	Final gas analysis O_2'	Final gas analysis P	Calcd. X	Calcd. A''	Calcd. O_2''
300	304	30	20.52	2.15	14.42	26.28	20.25	1.80
300	309	60	21.60	5.17	12.12	22.54	20.85	4.81
300	312	100	21.56	7.00	10.60	20.00	21.24	6.75
300	315	140	21.95	9.08	9.11	17.40	21.63	8.70
300	330	180	22.21	10.12	8.25	15.88	21.85	9.82
200	205	30	22.58	13.75	6.49	12.73	22.31	12.09
200	205	60	23.55	14.65	4.74	9.45	22.86	13.30
200	210	100	23.60	16.40	2.86	5.80	23.26	16.80
200	210	180	24.20	19.30	0.58	1.20	23.85	19.75
250	254	30	21.80	5.80	11.02	20.70	21.13	6.20
250	255	60	21.88	9.10	9.05	17.28	21.63	8.78
250	255	100	22.06	8.96	7.98	15.40	21.92	9.15
250	259	140	22.05	10.98	7.21	14.00	22.13	11.15
250	262	180	22.40	12.06	6.71	13.10	22.25	11.80
Gas mixture: (A) Ethylene, 21.00%; (O_2) Oxygen, 25.00%								
300	304	30	16.28	0.96	19.70	39.18	15.29	0.40
300	309	60	16.38	2.73	17.60	35.70	15.90	3.00
300	310	100	16.58	5.15	16.05	33.00	16.35	4.98
300	313	140	17.08	7.20	14.42	30.00	16.80	6.90
300	319	180	16.50	8.17	13.18	27.90	17.08	8.50
200	204	30	16.08	3.29	18.15	36.70	15.74	2.30
200	205	60	15.85	4.81	16.55	33.85	16.20	4.32
200	208	100	17.15	7.15	14.42	30.00	16.80	6.90
200	214	140	17.85	14.77	12.72	26.90	17.32	14.10
200	215	180	17.84	11.94	11.42	24.60	16.28	11.75
110	110	30	21.00	24.70	0.83	2.00	20.85	24.34
110	110	30	20.53	23.30	0.98	2.30	20.72	23.77

Defendant's Exhibit 80 (continued)

In the tables under the headings A', O_2' and F are given the results of the quantitative analysis of the gases after reaction. The column headed X gives the percentage of ethylene oxidized provided the reaction proceeds according to the following reaction: $C_2H_4 + 3O_2 \longrightarrow 2CO_2 + 2H_2O$. The fraction of ethylene oxidized is given by $x = 100\,f/(2A + 2Af)$, so that $100\,x = X$. From the amount of carbon dioxide formed it is also possible to calculate the amount of ethylene and oxygen which should be present with this carbon dioxide provided the oxidation of ethylene is complete. The calculated percentage of ethylene in the products of reaction is given by $A'' = A + f\,(A - 50)$, while for oxygen we have $O_2'' = O_2 + f\,(O_2 - 150)$. The values calculated for ethylene and oxygen check the experimental values very well and probably within the limits of the experimental error in the gas analysis.

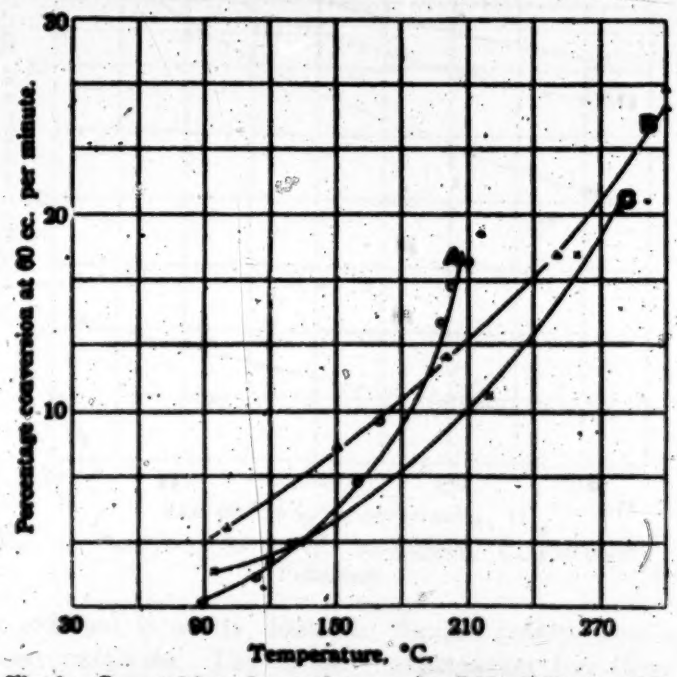


Fig. 1.—Composition of gas mixture: A = C_2H_4, 20%; O_2, 18%. B = C_2H_4, 21.3%; O_2, 16.85%. C = C_2H_4, 22.65%; O_2, 17.5%.

Discussion and Results

It is evident from these results that the catalysts used were effective in promoting the oxidation of ethylene at lower temperatures than previously reported. The copper, platinum and palladium catalysts began to be effective at about 100°. Fig. 1 shows the effect of temperature on the oxidation of ethylene for three different gas mixtures with platinized silica gel as the catalyst. The effect of increased rate of streaming on the con-

Defendant's Exhibit 80 (continued)

version is shown graphically in Fig. 2. The silver catalyst is shown to be less effective than the other catalysts. The catalysts all speeded up the successive oxidation reactions so that no intermediate products were detected. This confirms the previous work on the oxidation of ethylene.

The experimental results here given indicate that the reaction is directly proportional to the oxygen concentration and inversely proportional to the ethylene concentration. For example, when the ratio of ethylene to oxygen is changed from 1.36 to 0.81, the amount of ethylene oxidized by the platinum catalyst at 200° increases from 19.22 to 36.10%. Thus

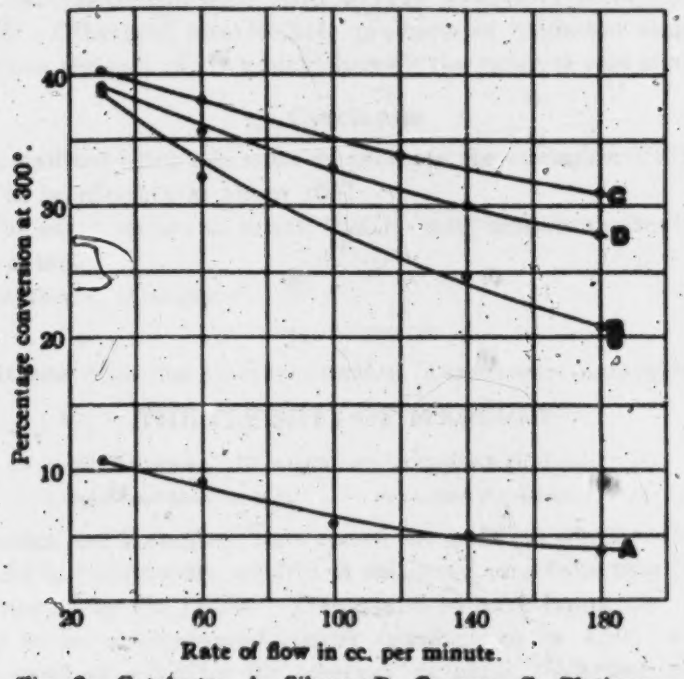


Fig. 2.—Catalysts: A, Silver; B, Copper; C, Platinum; D, Palladium.

the amount oxidized is nearly doubled. Similar results were obtained with the other catalysts. The simplest explanation for these results would seem to be that collisions of ethylene molecules with oxygen molecules which are adsorbed and activated by the effective catalyst centers resulted in reaction, whereas collisions of oxygen molecules with adsorbed ethylene molecules were ineffective. The adsorption studies of the authors[6] indicate specific adsorption of both ethylene and oxygen by these catalysts. Therefore, when the ethylene molecules are adsorbed by the active catalyst centers, then these centers are no longer effective. The double bond of ethylene is no doubt directed toward the catalyst. When,

[6] Reyerson and Swearingen, *J. Phys. Chem.*, 31, 88 (1927).

Defendant's Exhibit 80 (continued)

however, an oxygen molecule reaches an active center and is adsorbed, it is activated so that it is able to react with the ethylene which reaches it. Mere adsorption is not sufficient, otherwise the silica gel itself would be an active catalyst. Because of their higher energy content the primary products of oxidation are likely to be very reactive whether they are dihydroxyethylene or formaldehyde. Furthermore, as indicated in the tables, the temperature of the catalysts is often higher than that of the bath in which they are immersed. This indicates that the temperature at which the reaction takes place may be very much higher than that indicated, so that successive collisions with oxygen molecules result in complete oxidation. Otherwise intermediate products of oxidation should be detected when the rate of streaming through the catalyst was increased.

Conclusion

1. Metallized silica gels actively promote the oxidation of ethylene, beginning to be effective at about 100°.

2. The catalysts are so active that no intermediate products of oxidation are detected.

MINNEAPOLIS, MINNESOTA

[CONTRIBUTION FROM THE GAYLEY CHEMICAL LABORATORY, LAFAYETTE COLLEGE]

THE FLUIDITY OF MERCURY[1,2]

BY EUGENE C. BINGHAM AND THEODORE R. THOMPSON

RECEIVED JUNE 25, 1928 PUBLISHED NOVEMBER 6, 1928

Tammann and Hinnüber[3] have raised the question whether the viscosity of a liquid is trustworthy when it is measured in a tube that is not thoroughly wetted by the liquid. They claim to have found the viscosity of mercury in an amalgamated copper capillary to be 4.931 ± .78 *cp* at 13.5° instead of 1.609 *cp* for mercury in glass. Whether one accepts their explanation of the phenomenon based on "slippage," or not, the discrepancy of over 200%, is quite beyond experimental error and demands investigation.

1. The hypothesis of slippage is, of course, not new. It has often been offered to account for results which at the time seemed inexplicable, but on further investigation it has always been found that some factor has been overlooked which has made this explanation unnecessary. On the other hand, the fair agreement among the results of different investigators using widely different forms of viscometers and shearing stresses constitutes what may be fairly regarded as proof that slippage is certainly not

[1] In memory of Ira Remsen.

[2] Part of Master's Degree "Thesis" of Theodore R. Thompson.

[3] Tammann and Hinnüber, *Z. anorg. Chem.*, 167, 230 (1927).

Defendant's Exhibit 85.

XLVIII.—*On Oxide of Ethylene, considered as a link between Organic and Mineral Chemistry.*

[A Discourse delivered to the Fellows of the Chemical Society of London, June 5th, 1862.]

By M. Ad. Wurtz.

In the year 1795, four Dutch Chemists, Deiman, Troostwyk, Bondt, and Lauwerenburgh, first made the remarkable experiment of bringing together equal volumes of chlorine and olefiant gases, whereby they obtained a liquid compound, still known by the name of "Dutch liquid." This compound is an organic chloride, the dichloride of the diatomic radicle, olefiant gas or ethylene; and its constitution, or rather its mode of formation and most of its chemical properties, show that it may be represented by the formula

$$(C_2H_4)''Cl_2$$

Oxyde of ethylene is the corresponding oxide—

$$(C_2H_4)''O$$

This oxide cannot be formed directly by the action of oxygen on ethylene,* but it is obtained as a derivative of glycol, of which it is the anhydride or ether.

I must here draw attention to the re-actions by which glycol is produced, for they exhibit a remarkable peculiarity, on which I must especially insist. I obtained this body by the action of potash or baryta on diacetic glycol, a compound produced by the action of bromide or iodide of ethylene on acetate of silver, as shown by the following equation:—

$$(C_2H_4)''I_2 + \left\{\begin{matrix} C_2H_3O \\ Ag \end{matrix}\right\} O \Bigg\} + \Bigg\{ \begin{matrix} Ag \\ C_2H_3O \end{matrix}\Big\} O = 2AgI + \left\{\begin{matrix} C_2H_3O \\ C_2H_4 \\ C_2H_3O \end{matrix}\right\} O_2$$

Here we see clearly that 2 atoms of silver are removed, and their place supplied by ethylene, which, thus, as it were, rivets

* I have in vain endeavoured to bring about the direct combination of oxygen and ethylene, by heating a mixture of the two gases in the required proportions in a sealed flask containing acetic acid. I hoped to obtain by this method the acetate of ethylene or acetic glycol.

Defendant's Exhibit 86

N° 21,941 A.D. 1905

(Under International Convention.)

Date claimed for Patent under Patents Act, 1901, being date of first Foreign Application (in Germany), 27th Oct., 1904

Date of Application (in the United Kingdom), 27th Oct., 1905

Under Section 1 (2) of the Patents Act, 1901, this Specification became open to public inspection at the expiration of twelve months from the date of the application in Germany

Accepted, 27th Oct., 1906

COMPLETE SPECIFICATION.

Process for the Transformation of Organic Substances by Partial Combustion.

I, Doctor JOHANN WALTER, resident of 30 Chemin des Cottages, Geneva, Switzerland, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:

This invention relates to a process for the transformation of organic substances into others of greater commercial value by partial combustion substantially as hereinafter described.

Already in the year 1895, I published in the "Journal für Praktische Chemie" (New Series, vol. 51, page 107) an article entitled "Some experiments in oxidising by means of partial combustion": it was found however that the process there described could not be carried out profitably in practice. Since then I have continued my experiments, and the improved process which forms the subject-matter of the present application for patent is the result. I have now discovered the proper conditions under which the partial combustion must take place, which conditions are:

a) Thorough mixing of the gases, pulverized or vaporised substances to be treated with air, or in certain cases oxygen, as well as in some instances with steam, before they are conducted into the heating chamber. This mixing may be accomplished by means of a mechanically operated fan or any equivalent mixing apparatus.

b) Maintaining certain degrees of temperature in the heating chamber, which temperatures do not depend only upon the substance to be treated, but also on the speed of the current of gas or eventually current of steam, and on the cross-section of the heating chamber.

c) Mixing steam with the mixture of the substance to be partially burnt and air or, in certain cases, oxygen. The steam acts as a diluting agent,

 [*Price* 8*d.*]

Defendant's Exhibit 86 (continued)

2 N° 21,941.—A.D. 1905.

Process for the Transformation of Organic Substances by Partial Combustion.

prevents explosions, absorbs any excess of heat of combustion and facilitates the condensation of the transformed products.

d) Employment of the proper contact substance in those cases where such is necessary.

e) Employment of a combustion chamber of a cross section suited for the particular substance being treated, in certain cases the division or distribution of the well-mixed current of vapors and gas in a number of small heated tubes.

I will give a few instances of the applications of my invention:

Instance 1: As in the process of producing formic aldehyde according to the system of Loew-Tollens, as described in the "Berichte der Deutschen Chemischen Gesellschaft", 19th year, page 2133, so according to the process forming the object of the German Patent 55176 it is necessary to employ an excess of methyl alcohol in order to obtain a good yield of aldehyde, in relation to the alcohol consumed. The practical man knows this very well. The German Patent 106495 likewise draws attention to this point, and states moreover that this excess could be avoided by sprinkling methyl alcohol over large surfaces over which the air sweeps as a counter-current above the said alcohol; as also by adding nitrogen with the air. The nitrogen, however, not only uselessly increases the volume of incondensable gas and thus renders the condensation of the aldehyde difficult, but it has also been seen already, that the excess of methyl alcohol cannot thus be avoided, if it be incumbent to obtain as large a yield as possible. If on the contrary, instead of a gaseous means of dilution such as nitrogen or carbonic acid, steam be added, the object will be completely attained. It is true that the reaction $H.CH_2OH + O = H.COH + H_2O$ already gives 1 molecule of water but that is not enough to produce the favourable influence mentioned. Calculated in relation to the liquid of condensation, this molecule is approximately 37·3 *per cent* water. In order to obtain after cooling a solution of formic aldehyde of the usual degree, *viz.*, 10%, 22·7% of water may thus be added in the form of steam to the methyl alcohol vapours charged with air, before they are introduced into the heating chamber.

For the purpose of practically effecting the reaction, a copper vessel capable of holding about 100 litres is placed in a wooden cask, employed as a water bath. At the bottom of this vessel and inside the same is a copper coil perforated with small holes. The coil is continued outside and surrounds the vessel at a distance of about 50 mm and is likewise in the water bath. Through this coil a current of air is passed which can always be kept at the same pressure. The air is heated in the coil in the water-bath, passes out at the bottom of the copper vessel through the small openings in the coil, flows through the filling consisting of methyl alcohol and reaching to about ⅔ of the height, and becomes charged with vapour. The temperature of the water-bath is kept at 52° C by the introduction of steam. A thermometer situated in a pipe through which the mixture of methyl alcohol vapour and of air flows from the closed copper vessel, indicates 44° C. Into this pipe, behind the thermometer, a stream of controllable steam, issuing from a spraying nozzle, is introduced. This steam mixes with the stream of air carrying the methyl alcohol vapours. In looking further into the apparatus we find at a distance of about 10 mm apart two iron-wire pieces of net-work with fine meshes in the outflow pipe which separates behind the net-work into three short horizontal pipes each of which runs into a vertical copper pipe of about 80 mm diameter, containing a powder composed of asbestos and copper placed in thin layers on stout perforated copper plates.

At the beginning of each operation the asbestos and copper powder in each tube is heated to red heat by means of a blow-pipe applied to the outside of the tube at one of the places where the asbestos and copper touch the inside of the tube. The red heat begins to extend equally over the three layers and is self-maintained by the heat of combustion and is observed from

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Defendant's Exhibit 86 (continued)

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Process for the Transformation of Organic Substances by Partial Combustion.

time to time through mica doors placed above the layers. Below this the three pipes are joined again and form a single pipe, which enters a refrigerator with a pipe from which the condensed liquid, which causes an aerometer to float at some place in its course suitable for observation, flows into a collector, in which the aqueous solution of formic aldehyde is collected. The non-condensed vapours and gases before passing into the atmosphere flow through a small cylinder sprinkled with water and filled with pumice stones.

Instead of injecting air through the heated methyl alcohol the latter may be also distributed in the mixture of air and steam by means of a spraying nozzle or the methyl alcohol vapours separately prepared may be added to it. Further the pipe described in the communication as "a simple heating and refrigerating device for use in industry" which I have described in Part 2 of the "Chemische Industrie" for the year 1897 is heated internally with steam under pressure. A mixture of 4·3 parts of methyl alcohol and 1 part of water is injected to $^2/_3$ of the height of this pipe and air is admitted near A below in the external casing pipe whilst r is closed. In this case it is only necessary to regulate the air in order to obtain the determined proportion of mixture of air and methyl alcohol on the one hand and of steam on the other.

One volume of a natural gas consisting for the most part of methane (CH^4), thoroughly mixed with 5 volumes of air and 2 volumes of steam, is conducted into a tubular vessel closed at one end, and heated at the closed end to a powerful glowing heat, so that the relatively narrow supply pipe opens in the place where the heat is the greatest. Here the methane is consumed, the resulting product being formaldehyde and water. The vapours passing out of the heating chamber heat the fresh vapours flowing through the narrow middle tube, the first vapours hereupon entering a cooling device in which an aqueous solution of formaldehyde is deposited.

Instance III: A mineral oil distillate, boiling above 300° C., is sprayed into a current of air mixed with steam: this mixture is conducted over a contact substance consisting of asbestos coated with finely divided copper oxide, which is at a red heat, and then cooled in a condensing apparatus. By proper regulation of the proportions of the air and steam, the contact substance maintains itself at a red heat without further heat being applied outside. The product of condensation flowing out of the cooler has a strong acid reaction, the larger aqueous layer contains acetic acid and its homologues. From the oily layer floating at the top, an organic acid which solidifies at 0° C. can be separated by means of a soda solution and thereafter be precipitated by means of mineral acids and thereupon, by means of bisulphite of soda solution, an aldehyde which likewise solidifies and has a scent similar to peppermint. The principal quantity remaining is an oily fluid which begins to boil at 140° C. and of which up to 300° C. one half is distilled. The present process accordingly, like the so-called cracking process enables products of petroleum distillation boiling at a high temperature to be converted into products which boil at a low temperature.

Instance IV: In Beilstein's Handbook of Organic Chemistry 3rd Edition, vol. 1, page 112, it is stated: "An excess of ethylene with oxygen passed through a tube heated to 400° C. yields much trioxymethylene." It has not been possible heretofore to turn this reaction to practical account, owing to the excess of ethylene (C^2H^4), which has been lost. If only the theoretical combining quantities of ethylene and oxygen or air were taken, an explosion resulted. But if ethylene be mixed with the proper quantity of oxygen or air, and steam be then added thereto, and all thoroughly mixed before being passed into the heating chamber, the whole of the ethylene is recovered as formaldehyde or an aqueous solution of the same. In this case I find it preferable to form the heating chamber of a number of small narrow pipes.

Acetylene (C^2H^2) combines with hydrogen to form ethylene and consequently thereupon may be transformed by my process into formaldehyde. Ethylene

Defendant's Exhibit 86 (continued)

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Process for the Transformation of Organic Substances by Partial Combustion.

may also be obtained almost quantitatively from ethyl alcohol, either by G. S. Newth's method, *i.e.* dropping the ethyl alcohol on phosphoric acid heated to 200–220° C, or by W. Ipatiew's method, *i.e.* conducting the vapours of ethyl alcohol over heated aluminium oxide.

Instance V: In the manufacture of dimethylaniline from aniline, methyl alcohol and sulphuric acid, a gas consisting of methylether ($CH_3 \cdot O \cdot CH_3$) escapes on the cocks being opened after the cooling of the digesters. This gas has hitherto been allowed to escape into the atmosphere or it has been burnt up and thus transformed into carbonic acid and water. My process enables this gas to be turned to account by transforming it into formaldehyde. Manganous hydroxide is precipitated upon asbestos, then washed and dried and placed in a tube where it is heated to a low, barely noticeable red heat. As in the case of methane in Instance II, the methylether is thoroughly mixed with the necessary quantity of air and steam; and the mixture is then conducted over the manganous asbestos. On condensation of the vapors, an aqueous solution of formaldehyde is obtained.

This specification would run to too great a length were I to add more instances. I will only add that the reactions specified in Instances I, III, and V, by proper regulation of the temperature of the heating chamber, take place in the same manner but quantitatively with less favorable results, when no contact substance is employed; and on the other hand contact substances may also be employed in the reactions specified in Instances II and IV. The following may be employed as such contact substances: The metals platinum, iridium, palladium, silver, uranium, vanadium, iron, cobalt, nickel, cerium, thorium, copper, manganese, titanium, didymium, tungsten, also lead oxide by itself or mixed with lime, and chromoxide by itself or mixed with aluminium oxide or with lime—to which last mixture some potash may, in certain circumstances, be added. Molybdenic acid and thallium oxide may also be employed, but they volatilise readily, and they can only be recovered with difficulty. Arsenic and mercury may also be used as contact substances: they also volatilise, but their recovery is simple. In employing mercury for instance, some of it may be vaporised. The current of gas passing from the heating chamber carries quicksilver vapours with it: after partial cooling the current is conducted over rolls of copper or gold-wire net, on which the quicksilver forms an amalgam and is thus retained in the apparatus. By now reversing the direction of the current of gas and heating the amalgam, the quicksilver is given up again to the current, and after passing through the heating chamber it is absorbed by other copper rolls or equivalent, thus always remaining in the apparatus ready to play its part in the process.

Having now particularly described and ascertained the nature of my said invention and in what manner the same is to be performed, I declare that what I claim is:

1) A process for the transformation of organic substances to others of greater commercial value, by a partial combustion consisting in the condition that the partial combustion must take place under proper conditions which are: a° Thorough mixing by mechanical means, of the gas or substances to be transformed, with air, oxygen, or in certain cases with steam, before it enters the heating chamber; b° mixing with the above mixture, steam acting as diluting agent, c° maintaining particular temperature in the heating chamber.

2) A process for the transformation of organic substances into others of greater commercial value, by a partial combustion consisting in the condition that, in certain cases for the partial combustion as in Claim 1, the combustion must be carried out with contact substances and in other cases without any contact substances;

3) A process in accordance with Claim 1 for the transformation of organic substances into others of greater commercial value, by a partial combustion,

Defendant's Exhibit 86 (continued)

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Process for the Transformation of Organic Substances by Partial Combustion.

characterized by the condition, that in cases where a heating chamber of small section is necessary the mixed current of gas and vapor is distributed in a plurality of heating tubes of small inner diameter.

Dated this 27th day of October 1905.

WHEATLEY & MACKENZIE,
40, Chancery Lane, London, W.C.,
Agents.

Redhill: Printed for His Majesty's Stationery Office, by Love & Malcomson, Ltd.—1906.

DEFENDANT'S EXHIBIT 88

Liebig's Annalen der Chemie
1921, Vol. 422, pp. 36-46

III. The preparation of formaldehyde from ethylene

R. Willstätter & Max Bommer

* * *

Only at temperatures above 500°C. is ethylene rapidly oxidized by molecular oxygen. Under the experimental conditions of Bone and Wheeler its combustion proceeds at a very low velocity. With an ethylene oxygen mixture flowing through a glass tube with a time of heating of 1 second we observe at 410-450°C. only traces of a penetrating odor; with a mixture consisting of 60% ethylene, 5% of oxygen and 35% of nitrogen under the conditions of the experiments described further on a very faint odor of formaldehyde and a slight reddish discoloration of fuchsine-sulfurous acid appears at about 500°C.

It is obvious to lower the temperature of the slow combustion of ethylene by catalysts. However, whilst for the formation of formaldehyde from methyl alcohol, a dehydrogenation process, copper and silver are found to be serviceable, we find that for the oxidation of ethylene to formaldehyde accelerators of all kinds, viz. metals, such as osmium, platinum, silver and copper, as well as metal oxides, as for example those of cobalt, chromium, manganese and uranium, are utterly useless. The contact substances speed up the oxidation and lower the combustion temperature of the hydrocarbon, but they cause no formaldehyde to be formed. The unfavorable effect may either be due to the circumstance that they cause the action of oxygen on ethylene to proceed in another manner and lead to other oxidation products, or to the circumstance that they accelerate the dissociation of the formaldehyde even more than its formation.

It is already known that formaldehyde dissociates with great velocity into carbon monoxide and hydrogen even without any such contact action and already at a lower temperature than the one suitable for the preparation from ethylene. Thus W. A. Bone and H. L. Smith

described its decomposition from 400°C. onwards, and P. Sabatier has dealt in detail with the dissociation in the presence of metals and oxides.

As already shown by the investigation of F. C. Phillips on the oxidation of hydrogen and hydrocarbons, the combustion temperature is lowest in the presence of osmium. With ethylene we observed commencement of oxidation at 180°C. with direct formation of carbon dioxide; at 200°C. the oxygen was used up. With copper and silver likewise no formaldehyde was formed, and even with an excess of ethylene no carbon monoxide appears. The action of copper was already considerable at 250°C., at 400°C. and with a mixture of 80% of ethylene and 20% of oxygen half of the oxygen was consumed for forming carbon dioxide, and half of it by the copper.

On introducing a copper wire into the reaction space under the conditions most favorable to the formation of formaldehyde the formaldehyde disappears entirely; iron in a compact form, however, is harmless. • • •

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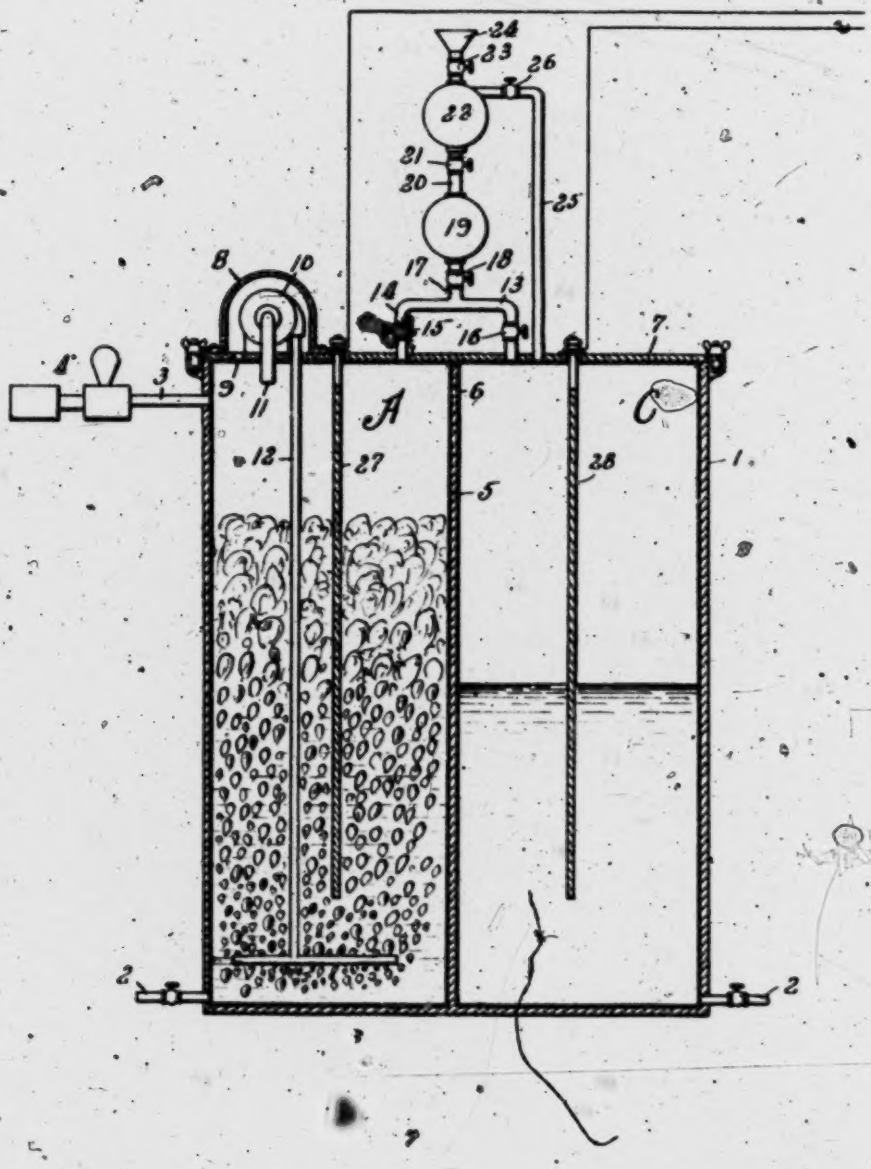
K. P. McELROY.

PROCESS OF AND APPARATUS FOR OXIDIZING HYDROCARBONS.

APPLICATION FILED AUG. 2, 1912. RENEWED MAR. 18, 1916.

1,253,617. Patented Jan. 15, 1918



Witnesses
H. C. Robinette
Dorsey Griffith

Inventor
K. P. McElroy

UNITED STATES PATENT OFFICE.

KARL P. McELROY, OF WASHINGTON, DISTRICT OF COLUMBIA, ASSIGNOR TO CHEMICAL DEVELOPMENT COMPANY, A CORPORATION OF MAINE.

PROCESS OF AND APPARATUS FOR OXIDIZING HYDROCARBONS.

1,253,617. Specification of Letters Patent. Patented Jan. 15, 1918.

Application filed August 2, 1912, Serial No. 712,975. Renewed March 18, 1916. Serial No. 85,200.

To all whom it may concern:

Be it known that I, KARL P. MCELROY, a citizen of the United States, residing at Washington, in the District of Columbia, have invented certain new and useful Improvements in Processes of and Apparatus for Oxidizing Hydrocarbons, of which the following is a specification.

This invention relates to processes of oxidizing hydrocarbons; and it comprises a method of producing useful products from ethylene and other gaseous and highly reactive unsaturated hydrocarbons wherein such a hydrocarbon is submitted to a direct limited oxidation in solution, the progress of the reaction being so controlled as to limit the extent of the oxidization to the production of simple oxygen-containing derivatives of such hydrocarbon and to preserve unattacked hydrocarbon in the sphere of reaction during the course of such reaction; all as more fully hereinafter described and as claimed.

There are various oxygen-containing or oxidized derivatives of ethylene, such as glycol, the chlorhydrin, glycol esters, glycol ethers, ethylene oxid, etc., having properties which would render them useful in various arts if such derivatives could be produced by a method sufficiently simple, cheap and ready to make them commercially available. At present however these bodies are not available for technical purposes being merely laboratory curiosities, made when wanted for scientific purposes by indirect, cumbrous and wasteful methods. Ordinarily, ethylene, made from alcohol, is converted into a dihalid and this is then saponified or esterified to replace the halogen by other groups. Saponification and esterification are both operations attended with some difficulty and giving small yields since the dihalids are stable, little reactive substances. Glycol, for example, is usually made for scientific purposes by converting ethylene, produced from alcohol, into the dibromid, converting the dibromid into an acetic ester and finally saponifying the latter by alkali.

It is an object of the present invention to obtain these potentially valuable oxidized bodies from ethylene and other olefinic gases by a direct limited oxidation under controlled conditions. Ethylene which is typical of the other olefinic gases is a readily oxidizable substance but being endothermically constituted, oxidation is attended with the liberation of much energy and is apt to be too far going, while the direct products of its limited oxidation, ethylene oxid, glycol and chlorhydrin are themselves also readily oxidizable. Ethylene oxid, which is the first product of oxidation, is particularly sensitive; glycol is more resistive.

I have found however that under suitable conditions the oxidation of ethylene can be limited and regulated to the desired degree and that such a limited oxidation of the ethlyene itself is, so to speak, preferential over the further oxidation of these proximate oxidation products; that is, so long as unattacked ethylene remains in the sphere of reaction the oxidation merely goes to the first stage and the ethylene exercises a protective action shielding the proximate products from further attack. In the present invention therefore conditions are so controlled as to restrain the violence of reaction, the heat of reaction being absorbed or removed as fast as evolved so as to maintain a predetermined temperature range; and an excess of ethylene is maintained in the sphere of reaction throughout the course of such reaction. While other methods of temperature control may be, and usefully are, employed dilution of the reacting bodies is desirable as this also aids in controlling the progress of the reaction. As stated, ethylene is endothermic and tends to enter into reaction with accelerating velocity; and dilution is useful in damping down the violence of the action.

Dilution is useful in the limited oxidation of ethylene in the gaseous state by chlorin and steam, steam being best present in ample amount, and by oxygen in the presence or absence of catalytics, such as platinum black, labile oxids, nitrous vapors, etc.; and in the treatment of ethylene dissolved in various liquids; advantageously an aqueous liquid. The solubility of ethylene in most aqueous liquids is relatively small so that even a saturated solution thereof is still actually, as regards the ethylene, very dilute; and the heat of oxidation becomes a limited quantity readily taken up and controlled by the liquid solvent. In fact the solubility of ethylene as ethylene in most aqueous liquids is so small as compared with that of many of the ordinary oxidants that in oxidizing in

aqueous solution special means must ordinarily be adopted to maintain the ethylene solution replenished with the gas and prevent the chance of an excess of such oxidant existing at any time during the reaction.

Several expedients to this end may be employed. One is an increase in pressure which causes a concomitant increase in the solubility of ethylene. With 15 pounds gage pressure, the amount which can be held in solution and drawn upon for reaction is about doubled. Another is quicker replenishment of the supply of dissolved ethylene by filming the liquid or otherwise increasing the area of contact between liquid and gas. Vigorous agitation of the liquid to prevent localized changes in the relative concentration of ethylene and oxidant is desirable. The increase in area of contact and the agitation may be simultaneously attained and a number of other desirable results secured by causing a cyclic circulation of the gas to, through and back to the reaction liquid; that is with a reaction chamber having a gas feed and means for withdrawal of waste gas there may be provided additional means capable of continuously taking gas from above the body of liquid and again bubbling it therethrough. Advantageously there may be employed a fan or blower. The fan means may be, and advantageously is, speeded up so as to maintain an emulsion of liquid and gas.

Another expedient often useful is to maintain porous, adsorptive materials, such as charcoal, coke, boneblack, platinum black, etc., suspended in the liquid in which the oxidant is contained or in which it is being developed to absorb gas and re-deliver it to the liquid. Such bodies may, and usually do, superadd a useful catalytic effect in aiding oxidation. For example, ethylene and air may be supplied to a hot liquid holding platinum black or colloidal palladium or the like in suspension.

Still another expedient is to maintain in contact with an aqueous reaction liquid or bath an oily liquid having a greater solvent power for ethylene to act, so to speak, as a store of ethylene. Oil of turpentine, for example, dissolves about 2½ times its volume of ethylene so that a drop of the oil saturated with a gas holds 2½ times as much ethylene as a bubble of the gas itself of the same size as such a drop. The use of an oily carrier of this kind is particularly advantageous with gases poor in ethylene, since it takes up ethylene and other unsaturated hydrocarbon gases with greater readiness than it does many other gases. In order to increase the surface of contact between the oily carrier and the aqueous oxidizing bath they should be kept emulsified together; and to promote ease of emulsification it is often useful to increase the specific gravity of the oil by mixing therewith heavy chlorinated oily bodies, such as chloroform, carbon tetrachlorid, Dutch liquid, etc., to an extent sufficient to make the specific gravities of the carrier and the bath at least approximately the same. And, vice versa, in using the heavy chlorinated products, a little petroleum hydrocarbon may be admixed to lighten them. However, with efficient stirring means the specific gravity of the carrier employed may not be important.

It is commonly useful to employ several of these expedients simultaneously as in feeding ethylene to an oxidizing chamber maintained under pressure as high as convenient, provided with means for cyclic gas circulation and containing in addition to the aqueous oxidizing bath an immiscible oily gas absorbent. However such expedients may be separately used.

The ethylene used in the present invention may be produced from any of the usual sources, as by dehydrating alcohol, by hydrogenating acetylene or by destructive distillation or cracking of coal, oil or other carbonaceous bodies. Using ordinary coal gas as a source of ethylene, it is useful to extract ethylene and other olefinic gases by means of wash oil; though the other oily carriers described may also be used. From the wash oil, the olefinic gases may be separated in a concentrated state by heating or vacuum. By proper precaution petroleum oil may be cracked down to form gas mixtures rich in ethylene, propylene and the butylenes; and useful in the present invention.

Acetylene may be hydrogenated to ethylene by catalytics or electrolytically.

Many oxidants may be used in the present invention, such as oxidizing salts, like permanganates, chromates, vanadates, arsenates, etc., in acid, alkaline or neutral baths. Peroxidized bodies, such as hydrogen peroxid, barium peroxid, calcium peroxid, sodium peroxid, perborates, persulfates, etc., may also be used. Manganese binoxid and an acid may be employed. Bleaching powder containing a trace of cobalt or other catalytic causing an evolution of nascent oxygen may be used. When ethylene chlorhydrin is desired, the bleaching powder may be acidulated with a weak acid. Or chlorin may be led into or formed in an aqueous solution of ethylene which may also contain neutralizing bodies, such as magnesia, chalk, soda, etc., to neutralize the HCl formed.

With many of these oxidants, the addition of the same or the development of its oxidizing influence should be proportionate to the rate of solution of ethylene; there should always be unattacked ethylene in the sphere of reaction.

It is advantageous to develop the oxidiz-

ing influence electrolytically, as this gives a better control of conditions and of the rate of development of such oxidizing influence. The rate of development can be readily adjusted to the rate of solution of the ethylene. Either diaphragmed or diaphragmless cells may be used. With a diaphragmed cell acetylene may be hydrogenated to ethylene in the cathode compartment and the ethylene oxidized in the anode compartment.

Any suitable electrolyte may be employed. With a saline solution, the alkaline cathode fluid may ultimately be used to neutralize the acid anode fluid to make a neutral solution from which the oxidation products may be regained. Any suitable anode metal and cathode metal may be used, and any suitable dissolved carrier such as titanium, cerium, manganese, iron, chromium, etc., salts may be used. Sulfate of soda is suitable for many purposes as a dissolved electrolyte. With salt (sodium chlorid) and other halogen compounds, the oxidation generally gives ethylene chlorhydrin which may be saponified to glycol with the soda accumulating in the cathode compartment. Ethylene chlorhydrin may be produced in any of the cells ordinarily employed for making hypochlorites, by using ethylene, or gases containing the same, as a depolarizer.

In the manufacture of caustic soda, it is an economical procedure to pass acetylene into the cathode compartment to act as a depolarizer and also produce ethylene, while the ethylene may be similarly used as a depolarizer in the anode compartment, producing either chlorhydrin or Dutch liquid as the case may be. In this way the consumption of energy for the production of caustic soda may be much lessened while useful products are obtained to aid in paying the expense of the operation.

In the accompanying illustration I have shown, more or less diagrammatically, a generalized type of apparatus which may be used in many of the reactions described herein. In this showing

Reference character 1 indicates a vessel which may be of any suitable material, such as sheet steel, appropriately coated with a resistant inner lining. This is advantageously built so as to withstand internal pressure. It is provided with valved draw-off outlets 2 for removing liquid. It is also provided with inlet 3, supplied by pressure pump 4, through which an olefinic gas, or mixtures of such gas with any other gas desired, may be supplied under any convenient pressure. As shown, the vessel is particularly adapted for electrolytic purposes and is provided with diaphragm 5 of any usual material dividing it into an anode chamber A and cathode chamber C. This diaphragm is advantageously perforated at 6 to allow equalization of gas pressure on the two sides. The vessel is provided with cover 7 which may be clamped on in any suitable manner. This cover carries hood 8 having communication with the chamber of the vessel through orifice 9. This equalizes the chamber pressure with that of the hood 8. Within this hood is mounted any simple type of blower or pump, shown here as rotary fan 10. This blower has communication with the gas space within the chamber at 11 and is provided with conduit 12 extending nearly to the bottom of the vessel. The lid may also be provided with two conduits 13 and 14 placed on each side of the diaphragm and valved respectively at 15 and 16. These two conduits unite as conduit 17, valved at 18, which expands into chamber 19. Above this chamber is a length of tubing 20, valved at 21, and connecting with another chamber 22, above which is valve 23 and hopper or funnel 24. Equalizing pipe 25, valved at 26, allows equalization of pressure. This last described device enables equalization of gas pressure with that of the chamber by opening appropriate valves; and it also allows liquid to be fed to either side of the diaphragm and without disturbance of the chamber pressure. For example, a liquid may be fed into 24, and, valves 26 and 21 being closed, may be fed into 22 by opening valve 23. By now closing valve 23 and opening 26 and 21, (valve 18 being normally open) the liquid may be caused to pass down into 19 and thence by 16 or 15, the liquid may be discharged on either side of the diaphragm.

Anode 27 and cathode 28 may be of any suitable metal or structure adapted to a particular electrolysis.

In one embodiment of the present invention, using electrolytic oxidation, the vessel may be charged on both sides of the diaphragm with a solution of sulfate of soda of such strength as to give good conduction. Ethylene, or gas containing ethylene, may now be introduced into the apparatus by means of pump 4 at any pressure desired, say, 4 or 5 atmospheres. Upon now starting blower 10 into operation, the liquid may be kept thoroughly saturated with dissolved ethylene. It is advantageous to run this pump or blower at such a rate that the liquid is maintained as more or less of an emulsion of gas and liquid; as a foamy mass. Upon now passing a current through the device in such a manner that 27 becomes the anode, the ethylene is oxidized to glycol which goes into solution. Nascent oxygen, ozone, hydrogen peroxid, etc., produce at the anode an oxidation of ethylene in the presence of water to form glycol. The rate of development of the oxygen should be limited according to the rate of solution of ethylene so that an excess of ethylene shall remain in the solution at all times. Should the solu-

tion temporarily become exhausted of ethylene, there would be danger of the oxidation of glycol to form glycolic acid, etc. For this reason the current should be regulated in correspondance with the supply of dissolved ethylene. The apparatus may be run discontinuously, a charge of ethylene, or gas containing the same, as for example, a pyrolytic oil gas containing say 40 to 50 per cent. of ethylene and other olefins, introduced and the pump 10 kept in operation until the ethylene of the charge has become absorbed and oxidized. At this time the current may be interrupted and waste gas vented through 14. Another charge of ethylene, or gas containing the same, may now be introduced and so on until the concentration of glycol in the solution becomes as high as may be desired. In the electrolytic operation using sodium sulfate as the conductive electrolyte, alkali of course is developed in the cathode compartment in amount corresponding to the amount of acid in the anode compartment. At the end of the operation, by uniting the anode fluid and the cathode fluid, a neutral saline solution is regained and from this glycol may be isolated in ways similar to the ways of recovering glycerin from salt lyes its properties, as well as its uses, being very similar to those of glycerin. Like glycerin it is a dense, high boiling liquid which however can be blown over readily with steam. The anode may be of platinum, carbon or lead. The anode fluid may contain a carrier oxidant, such as a chrominum salt, a cerium salt, a manganese salt, etc. Other oxygen salts, such as phosphates, borates, etc., may be used in lieu of the sulfate, but the sulfates are advantageous. Sulfate of soda is a good sulfate.

In the anode chamber may be maintained a little suspended platinum black, charcoal, boneblack, etc., if desired. It may also contain an oil having solvent properties for ethylene, such as turpentine, mineral oil, etc. With proper manipulation and the maintenance of dissolved ethylene in the solution at all times, oxidation will be restricted to the ethylene.

By substituting a solution of common salt in the cell for the solution of sulfate of soda just described but otherwise operating in the same manner, the reaction with oil gas will produce ethylene chlorhydrin and other chlorhydrins. In this action since chlorin is withdrawn from the sphere of reaction in the form of chlorhydrin which is a neutral body, the alkalinity of the cathode fluid increases more rapidly than the acidity of the anode liquid. The operation may be continued until the per cent. of salt converted into alkali and chlorhydrin is as high as may be desired, the liquid being replenished with salt from time to time if desired. Operating in this manner the process becomes the manufacture of caustic soda with the use of ethylene as a depolarizer to reduce the current consumption and with the production of a valuable byproduct, the chlorhydrin, in lieu of chlorin. Ethylene chlorhydrin is valuable as a solvent in addition to its value as a material for conversion into glycol.

In case it is desired to form glycol from the chlorhydrin, the anode fluid and the cathode fluid may be united and heated with such chlorhydrin under pressure. Without pressure the action of caustic alkali on the chlorhydrin is to convert it into the corresponding oxid. The alkalinity of the cathode fluid is sufficient to neutralize the acidity of the anode fluid and also to hydrolyze the chlorhydrin which has been formed.

In using salt as the electrolyte, the diaphragm may be dispensed with and the cell worked under conditions analogous to those employed in making hypochlorites. Operating in this way, the cathode should be of small effective area to give a high current density and permit the escape of hydrogen. For this latter purpose, it may be provided with a conduit for taking away hydrogen. The ethylene being present to unite with the chlorin and with hydroxyl, the cell does not produce hypochlorites as long as it remains in excess.

In working with salt as an electrolyte, it is desirable to keep the temperatures low and to maintain the stated excess of ethylene present. In the absence of an excess of dissolved ethylene, if the electrolysis be pushed, chlorin formed as such may combine with gaseous ethylene to form Dutch liquid. This however is a valuable material.

Operating with pure ethylene, such as that made by the hydrogenation of acetylene, in the cell having a dissolved oxygen salt, pure glycol will be formed. But with pyrolytic gases containing ethylene, such as those obtained by cracking oils at low temperatures, there will usually be propylene and the butylenes present. These also make valuable glycols. Using pure ethylene in a cell having a chlorid as the electrolyte, similarly pure chlorhydrin, or chlorhydrin and Dutch liquid, will be produced. But with pyrolytic gases containing other olefins than ethylene, other chlorhydrins and chlorids may be obtained.

The present process may be used in connection with pure propylene or the butylenes in the manufacture of propylene glycol and propylene chlorhydrins, butylene glycols and butylene chlorhydrins.

In one modification of my invention, I may mount a gas circulating device on the cathode chamber C as well as on the anode chamber A, and in the former I may circulate acetylene. The acetylene acts as a depolarizer, combining with hydrogen to form

ethylene and reducing the current consumption. This ethylene may now be transferred to the anode chamber and oxidized to form glycol or chlorhydrin as the case may be. Running a cell for the production of caustic soda from brine in this manner, the current consumption is much reduced. In hydrogenating acetylene in this manner, it is useful to employ cathodes of nickel, cobalt and the like.

Apparatus of the type described and shown may also be used for non-electrolytic methods under the present invention. By omitting diaphragm 5, anode 27 and cathode 28, an oxidizing liquid may be fed in at a regulated rate through funnel 24 and connected parts. This oxidizing liquid may be a milk of bleaching powder. The rate of feed should be such as not to exceed the rate of solution of ethylene in the fluid of the chamber. Potassium permanganate solution may be fed in in the same manner. Many other oxidizing fluids may be similarly used. Or a charge of bleaching powder and water may be fed in through 24 and connected parts and then a mixture of carbon dioxid and ethylene fed in by pump 4. The carbon dioxid will decompose the bleaching powder forming hypochlorous acid which will unite with the ethylene to form chlorhydrin. Or the apparatus may be charged with bleaching powder and water containing a very small proportion of a cobalt salt and ethylene fed in and circulated as before. In the presence of a cobalt salt as a catalyst, bleaching powder gives up oxygen at a slow rate; and this oxygen will oxidize the ethylene. The rate may be controlled by controlling the amount of cobalt salt present and should be such that the rate of development of oxygen will not exceed the rate of solution of ethylene.

In another modification, the apparatus may be maintained at any suitable pressure and temperature and a mixture of ethylene and oxygen, or air, fed in and circulated, the liquid being kept charged with suspended or dissolved catalytic, such as platinum black, palladium, alkaline manganate, etc. The ethylene should be in excess of the oxygen at all times.

Using the rocker type of cell in electrolyzing brine, acetylene may be hydrogenated by contacting with the mercury in the soda compartment and the ethylene produced used as an anodic depolarizer in forming chlorhydrin.

Gaseous acetylenic hydrocarbons, such as acetylene, and gaseous diolefinic hydrocarbons, as well as gaseous olefins, may be advantageously oxidized by the present process.

In using weak ethylenic gases, such as those from cracking oil, from acetylene hydrogenated by nickel in the presence of a diluent gas, etc., if operating with a single electrolytic cell discontinuously, that is, with successive charges of gas introduced and circulated until the ethylene is absorbed, there is a periodic variation in the strength of current required. For example, with a fresh charge of coal gas containing, say, 45 per cent. ethylene and other olefins with the residue largely ethane, the absorption of the gas is at first relatively rapid, but as the ethylene disappears solution becomes slower; being for instance considerably slower when the richness in ethylene is reduced to 30 per cent. This may need a corresponding change in the current supply. By operating a number of cells in series, as regards the gas feed, each such cell venting gas into the next in series, this inconvenience may be overcome, each cell then operating with gases of a constant average content; the first, say with gas averaging 40 per cent., the next with gas averaging 30 per cent., and so on, each cell then having a constant current supply.

Gas of any desired degree of purity may however be obtained from lean ethylenic gas by scrubbing the latter by a countercurrent of spirits of turpentine, heavy oil or any other solvent for ethylene, exhausting the charged solvent by vacuum or heat, or both, and returning the exhausted solvent for fresh use with the gas to be scrubbed. The rich ethylene obtained by exhausting the charged oil may then be oxidized.

The manufacture of chlorhydrin is not specifically claimed in this case, it forming part of the matter of certain later but copending applications Serial Nos. 34,678, filed June 17, 1915; 35,505, filed June 22, 1915; 35,646, filed June 22, 1915; 65,983, filed Dec. 9, 1915; 107,564, filed July 5, 1916; 147,465, filed Feb. 8, 1917.

What I claim is:—

1. The process of preparing useful products which comprises submitting an unsaturated gaseous hydrocarbon to limited oxidation, such hydrocarbon being maintained in excess in the sphere of reaction during such oxidation.

2. The process of preparing useful products which comprises submitting a gaseous olefinic hydrocarbon to limited oxidation, such hydrocarbon being maintained in excess in the sphere of reaction during such oxidation.

3. The process of preparing useful products which comprises submitting a gas comprising ethylene to limited oxidation, such ethylene being maintained in excess in the sphere of reaction during such oxidation.

4. The process of making useful products which comprises subjecting a gaseous unsaturated hydrocarbon to limited anodic oxidation, such gaseous hydrocarbon being maintained in excess in the sphere of reaction during such oxidation.

5. The process of making useful products which comprises subjecting a gaseous olefinic hydrocarbon to limited anodic oxidation.

6. The process of making useful products which comprises subjecting a gas comprising ethylene to limited anodic oxidation.

7. The process of making useful products which comprises subjecting a gaseous unsaturated hydrocarbon to limited anodic oxidation with a sulfate electrolyte, such gaseous hydrocarbon being maintained in excess in the sphere of reaction during such oxidation.

8. The process of making useful products which comprises subjecting a gaseous olefinic hydrocarbon to limited anodic oxidation with a sulfate electrolyte.

9. The process of making useful products which comprises subjecting a gas comprising ethylene to limited oxidation with a sulfate electrolyte.

10. The process of making useful products which comprises electrolyzing sodium sulfate solution in the presence of a gaseous unsaturated hydrocarbon as a depolarizer.

11. The process of making useful products which comprises electrolyzing sodium sulfate solution in the presence of a gaseous unsaturated hydrocarbon as an anodic depolarizer.

12. The process of making useful products which comprises electrolyzing sodium sulfate solution in the presence of a gaseous olefinic hydrocarbon as an anodic depolarizer.

13. The process of making useful products which comprises electrolyzing sodium sulfate solution in the presence of a gas comprising ethylene as an anodic depolarizer.

14. In the electrolysis of solution of salts, the process which comprises electrolyzing a solution of a suitable salt in a divided cell, hydrogenating acetylene in the cathode compartment to produce ethylene and transferring the ethylene to the anode compartment to act as a depolarizer and produce chlorhydrin.

15. In the oxidation of unsaturated gaseous hydrocarbons in aqueous solution, the process which comprises maintaining in contact with such solution an oily solvent for such hydrocarbon while passing a current of such hydrocarbon through such solution.

16. In the oxidation of unsaturated gaseous hydrocarbons in aqueous solution, the process which comprises maintaining in contact with such solution a substance having the power of physically absorbing such hydrocarbon while passing a current of such hydrocarbon through such solution.

17. In the anodic oxidation of gaseous hydrocarbons, the process which comprises establishing an electrolytic current through a suitable electrolyte, and transmitting a current of such gaseous hydrocarbon through the electrolyte while maintaining in contact with said electrolyte an oily fluid having a solvent power for said gaseous hydrocarbon.

18. In the oxidation of ethylene, the process which comprises supplying ethylene gas under pressure to an aqueous liquid wherein oxidizing conditions are maintained in such manner as to maintain an excess of ethylene in the sphere of reaction at all times.

19. In the oxidation of ethylene the process which comprises supplying gas comprising ethylene under pressure to a closed chamber containing a body of aqueous liquid wherein oxidizing conditions are maintained, and causing a cyclic circulation of the gas so supplied through said body of liquid.

20. In the oxidation of olefinic gases, the process which comprises progressively developing oxidizing influences in a body of aqueous liquid and supplying dissolved olefinic gas to such liquid at a rate sufficiently rapid to maintain an excess of dissolved gas at all times during the oxidation.

21. In the oxidation of ethylene, the process which comprises progressively developing oxidizing influences in a body of aqueous liquid and supplying dissolved ethylene to such liquid at a rate sufficiently rapid to maintain an excess of dissolved ethylene at all times during oxidation.

22. In the treatment of difficultly soluble gases in solution, the process which comprises establishing reactive conditions in a body of liquid in a suitable chamber, supplying gas to be treated to said chamber and establishing a cyclic circulation of said gas independent of the supply thereof from a point above the liquid body to and through such body.

23. In the oxidative treatment of difficultly soluble gases in solution, the process which comprises establishing reactive oxidizing conditions in a body of liquid in a suitable chamber, supplying gas to be treated to said chamber and establishing a cyclic circulation of said gas independent of the supply thereof from a point above the liquid body to and through said body, said circulation being at a sufficiently rapid rate to maintain said body vesicular with said gas.

24. In the depolarization of electrolytic cells, the process which comprises supplying a feed of depolarizing gas to such a cell while maintaining an independent cyclic circulation of said gas through liquid in said cell, said circulation being sufficiently rapid to maintain said liquid in a foamy state.

25. In the manufacture of solvents, the process which comprises electrolyzing a liquid with a suitable anode and cathode,

supplying a feed of gas comprising a gaseous olefin and producing a rapid cyclic circulation of such gas through the electrolyzing fluid in the vicinity of the anode.

26. In the manufacture of solvents, the process which comprises electrolyzing a liquid with a suitable anode and cathode, supplying a feed of gas comprising ethylene and producing a rapid cyclic circulation of such gas through the electrolyzing fluid in the vicinity of the anode.

27. In oxidizing apparatus, a closed vessel, means for electrolyzing liquid therein, means for supplying gas thereto, means for removing waste gas therefrom and means for producing an independent cyclic circulation to gas within such vessel.

28. In oxidizing apparatus, a closed vessel, means for electrolyzing liquid therein, means for supplying gas thereto, means for removing waste gas therefrom and means for producing an independent cyclic circulation of gas within such vessel, such means comprising a fan having an inlet in the top of said vessel and an outlet conduit terminating in its base.

29. In an electrolytic apparatus, a closed cell provided with an anode and a cathode, means for feeding gas to said cell, means for withdrawing waste gas from such cell and independent means for producing cyclic circulation of gas within such cell.

30. In an electrolytic apparatus, a closed cell provided with an anode and a cathode, means for feeding gas to said cell, means for withdrawing waste gas from such cell and independent means for producing cyclic circulation of gas within such cell, such means comprising a fan having an inlet in the top of said cell and an outlet conduit terminating in its base.

31. In the manufacture of useful products, the process which comprises submitting an unsaturated hydrocarbon to limited oxidation, and during said oxidation and while supplying hydrocarbon to the reaction chamber continuously maintaining therein oxidizing conditions.

32. The process of manufacturing useful products which comprises producing a gaseous mixture containing an olefin and treating said mixture to produce an addition compound of said olefin containing at least one alcohol radical.

33. The process which comprises producing a gaseous mixture containing a gaseous olefin and flowing said gas in intimate contact with an aqueous solution capable of delivering an alcohol radical, whereby a substantial proportion of the olefin reacts with a portion of said solution to form an addition compound containing at least one alcohol radical.

In testimony whereof, I affix my signature in the presence of two subscribing witnesses.

K. P. McELROY.

Witnesses:
JOHN H. SIGGERS,
H. SCHOENTHAL.

K. P. McELROY.

PROCESS OF OXIDIZING HYDROCARBONS.

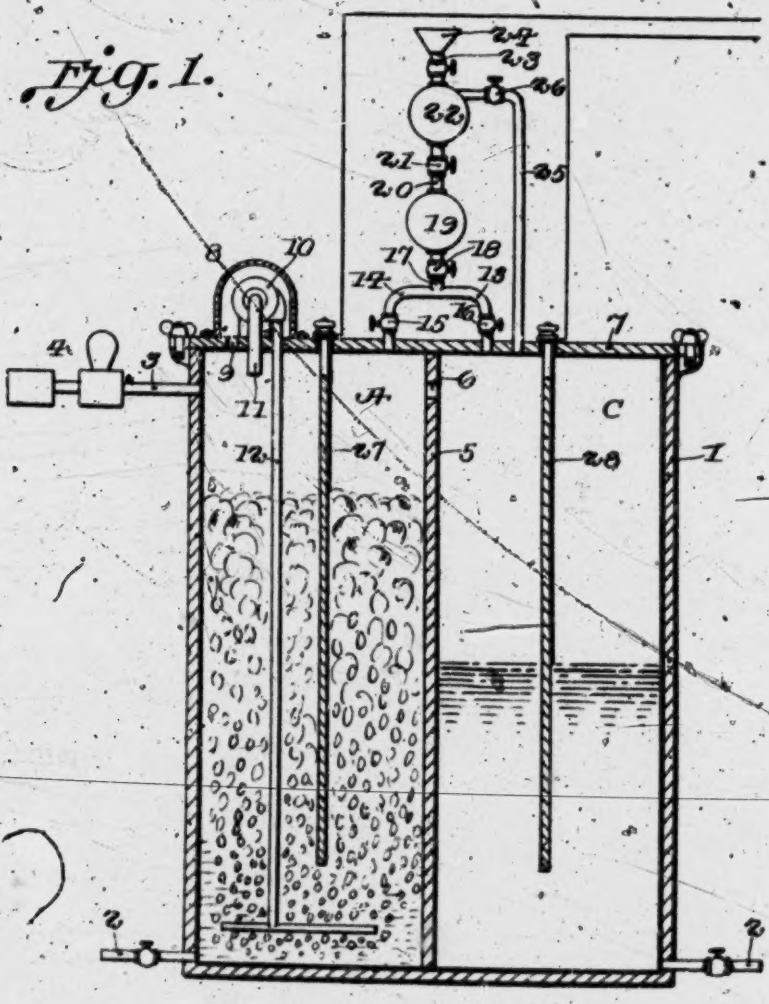
APPLICATION FILED JUNE 26, 1917.

1,308,797.

Patented July 8, 1919.

2 SHEETS—SHEET 1.

Fig. 1.



Witness

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PROCESS OF OXIDIZING HYDROCARBONS.

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1,308,797.

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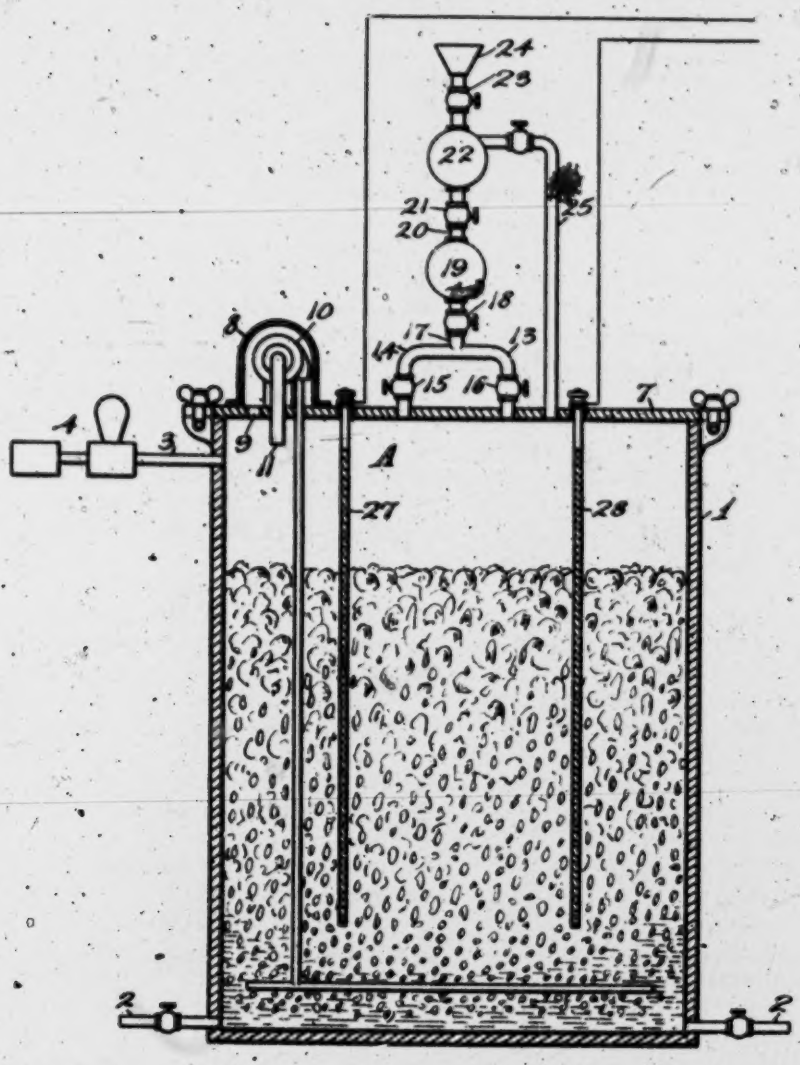
2 SHEETS—SHEET 2.

Fig. 2.



Inventor:
Karl P. McElroy
By Philipp Sawyer Rule & Kennedy

UNITED STATES PATENT OFFICE.

KARL P. McELROY, OF WASHINGTON, DISTRICT OF COLUMBIA, ASSIGNOR TO CHEMICAL DEVELOPMENT COMPANY, A CORPORATION OF MAINE.

PROCESS OF OXIDIZING HYDROCARBONS.

1,308,797. Specification of Letters Patent. Patented July 8, 1919.

Application filed June 26 1917. Serial No. 177,028.

To all whom it may concern:

Be it known that I, KARL P. McELROY, a citizen of the United States, residing at Washington, in the District of Columbia, have invented certain new and useful Improvements in Processes of Oxidizing Hydrocarbons, of which the following is a specification.

This invention relates to processes of oxidizing hydrocarbons, and it comprises a method of producing useful products from unsaturated hydrocarbons, and particularly from olefin hydrocarbons, wherein such a hydrocarobon is submitted to direct oxidation in solution with the aid of a catalyst, and particularly a metal of the platinum group, oxygen for oxidation being supplied in the form of an oxidizing salt or other suitable source of oxidation.

There are various oxygen-containing or oxidized derivatives of the olefins, such as the glycols, the chlorhydrin, glycol esters, glycol ethers, olefin oxids, etc., having properties which would render them useful in various arts if such derivatives could be produced by a method sufficiently simple, cheap and ready to make them commercially available. At present however these bodies are not available for technical purposes being merely laboratory curiosities, made when wanted for scientific purposes by indirect, cumbrous and wasteful methods. For example, ethylene, ordinarily made from alcohol, is converted into a dihalid and this is then saponified or esterified to replace the halogen by other groups. Saponification and esterification are both operations attended with some difficulty and giving small yields since the dihalids are stable, little reactive substances. Ethylene glycol, for example, is usually made for scientific purposes by converting ethylene, produced from alcohol, into the dibromid, converting the dibromid into an acetic ester and finally saponifying the latter by alkali.

It is an object of the present invention to obtain these potentially valuable oxidized bodies from ethylene and other olefins in the gaseous condition by a direct limited oxidation under controlled conditions. Ethylene which is typical of the other olefinic gases is a readily oxidizable substance, but, being endothermerically constituted, oxidation is attended with the liberation of much energy and is apt to be too far going, and furthermore the direct products of its limited oxidation, ethylene oxid, glycol and chlorhydrin are themselves also readily oxidizable. Ethoylene oxid, which is the first product of oxidation, is particularly sensitive; glycol is more resistive.

I have found however that under suitable conditions the oxidation of the olefins, for example, ethylene can be limited and regulated to the desired degree and that such a limited oxidation of an olefin such as ethylene itself is, so to speak, preferential over the further oxidation of these proximate oxidation products; that is, so long as unattacked ethylene remains in the sphere of reaction the oxidation merely goes to the first stage and the ethylene exercises a protective action shielding the proximate products from further attack.

Dilution is useful in the limited oxidation of ethylene in the gaseous state by chlorin and steam, steam being best present in ample amount, and by oxygen in the presence or absence of catalytics, such as platinum black, labile oxids, nitrous vapors, etc.; and in the treatment of ethylene dissolved in various liquids; advantageously an aqueous liquid. The solubility of ethylene in most aqueous liquids is relatively small so that even a saturated solution thereof is still actually, as regards the ethylene, very dilute and the heat of oxidation becomes a limited quantity readily taken up and controlled by the liquid solvent. In fact the solubility of ethylene as ethylene in most aqueous liquids is so small as compared with that of many of the ordinary oxidants that in oxidizing in aqueous solutions special means must ordinarily be adopted to maintain the ethylene solution replenished with the gas and prevent the chance of an excess of such oxidant existing at any time during the reaction.

Several expedients to this end may be employed. One is an increase in pressure which causes a concomitant increase in the solubility of ethylene. With 15 pounds gage pressure, the amount which can be held in solution and drawn upon for reaction is about doubled. Another is quicker replenishment of the supply of dissolved ethylene by filming the liquid or otherwise increasing the area of contact between liquid and gas. Vigorous agitation of the liquid to prevent localized changes in the relative concentration of ethylene and oxidant is desirable. The

increase in area of contact and the agitation may be simultaneously attained and a number of other desirable results secured by causing a cyclic circulation of the gas to, through and back to the reaction liquid; that is with a reaction chamber having a gas feed and means for withdrawal of waste gas there may be provided additional means capable of continuously taking gas from above the body of liquid and again bubbling it therethrough. Advantageously there may be employed a fan or blower. The fan means may be, and advantageously is, speeded up so as to maintain an emulsion of liquid and gas.

Another expedient often useful is to maintain porous, absorptive materials, such as charcoal, coke, boneblack, platinum black, etc., suspended in the liquid in which the oxidant is contained or in which it is being developed to absorb gas and re-deliver it to the liquid. Such bodies may, and usually do, superadd a useful catalytic effect in aiding oxidation. For example, ethylene and air may be supplied to a hot liquid holding platinum black or colloidal palladium or the like in suspension.

Where the unsaturated hydrocarbon is ethylene it may be produced from any of the usual sources, as by dehydrating alcohol, by hydrogenating acetylene or by destructive distillation or cracking of coal, oil or other carbonaceous bodies. Using ordinary coal gas as a source of olefins, such as ethylene, it is useful to extract the ethylene and other olefinic gases by means of wash oil. From the wash oil, the olefinic gases may be separated in a concentrated state by heating or vacuum.

By proper precaution petroleum oil may be cracked down to form gas mixtures rich in ethylene, propylene and the butylenes; and useful in the present invention. Acetylene may be hydrogenated to ethylene by catalytics or electrolytically.

Many oxidants may be used in the present invention, such as oxidizing salts, like permanganates, chromates, vanadates, arsenates, etc., in acid, alkaline or neutral baths. Peroxidized bodies, such as hydrogen peroxid, barium peroxid, calcium peroxid, sodium peroxid, perborates, persulfates, etc., may also be used. Manganese binoxid and an acid may be employed. Bleaching powder containing a trace of cobalt or other catalytic causing an evolution of nascent oxygen may be used. When ethylene chlorhydrin is desired, the bleaching powder may be acidulated with a weak acid.

With many of these oxidants, the addition of the same or the development of its oxidizing influence should be proportionate to the rate of solution of ethylene; there should always be unattacked ethylene in the sphere of reaction.

The drawings illustrate, diagrammatically, apparatus which is suitable for carrying out the invention. Figure 1 shows apparatus with a diaphragm and Fig. 2 shows another type of apparatus not employing a diaphragm.

The invention is particularly intended to be carried out in an electrolytic cell, and in this case any suitable electrolyte may be used, such as salt (sodium chlorid), sodium sulfate, phosphates, borates, etc. Electrolysis may be in a divided cell, as shown in Fig. 1, that is a cell having a diaphragm separating the cell into an anode compartment and a cathode compartment; or in an undivided or diaphragmless cell, such as those used in electrolyzing sodium chlorid solutions to make hypochlorites and other oxidized chlorin-containing products, as shown in Fig. 2. The results differ with the type of cell. With a divided cell, electrolysis necessarily develops oxidizing conditions and acidity in the anode chamber and reducing conditions and basicity in the cathode chamber. In such a cell using a chlorid electrolyte, part of the chlorin developed goes into combination with the ethylene, etc., to form chlorhydrins. Using such a cell, the anode liquor and cathode liquor may be ultimately reunited to form a neutral liquid from which the oxidation products of the ethylene, etc., may be regained. With an undivided cell, the electrolyte remains neutral but oxidizing conditions develop while hydrogen escapes. If the electrolyte contains an oxygen salt such as a sulfate, borate, etc., the oxygen mostly develops as such and for economy it must be used (that is taken up by the olefin) as fast as it is produced since these salts themselves do not ordinarily oxidize to any great extent; that is there is not much tendency, except under special conditions, to form persulfates and the like. The use of carrier oxidants, such as chromium salts, iron salts, cerium salts, manganese salts, etc., however may be resorted to since such salts are able to take up and redeliver oxygen. With a chlorid salt, such as sodium chlorid, in an undivided cell, the circumstances are different since the net effect of development of oxidation is then a production of oxidizing salts such as hypochlorites and chlorates. These are oxidizing salts which may be used to redeliver oxygen with the aid of suitable catalysts. Therefore any oxygen not at once taken up by the olefin may be, so to speak, stored. With an undivided cell the solution, as stated, remains neutral and these oxidized chlorin products have, in neutral solution, no violent oxidizing action. Any suitable anode metal and cathode metal may be used.

With salt (sodium chlorid) and other halogen compounds in a divided cell, on oxidizing ethylene in the anode chamber, the product is generally ethylene chlorhydrin

which may be saponified to glycol with the soda accumulating in the cathode compartment. In forming chlorhydrin, chlorin goes into organic combination and the net result is the formation and accumulation in the cathode chamber of an excess of alkali corresponding to the amount of chlorin so removed. On saponification (removal of the chlorin from organic combination), with this excess of alkali a neutral liquor is once more formed.

In electrolyzing a salt solution in an undivided cell in the usual ways as stated the salt may be in effect oxidized with formation of hypochlorites and chlorates but the neutrality of the solution does not change. If this electrolysis be performed in the presence of olefins, etc., the net result is the formation of glycols; and it is immaterial here whether this result is a direct oxidation of the ethylene in preference to the salt, is a preliminary oxidation of the salt to hypochlorite, etc., with transference of oxygen from the oxidized salt to the ethylene, or is a preliminary formation of chlorhydrin with subsequent saponification to glycol by the alkali simultaneously formed. In working with the undivided cell and a sodium chlorid solution it is better to use oxygen carrying catalysts, such as cobalt salts, or platinum group metals, such as platinum, palladium, etc.

In the accompanying illustration I have shown, more or less diagrammatically, a generalized type of apparatus which may be used in many of the reactions described herein. In this showing reference character 1 indicates a vessel which may be made of any suitable materials, such as sheet metal, appropriately coated with a resistant inner lining. This is advantageously built so as to withstand internal pressure. It is provided with valved draw-off outlets 2 for removing liquid. It is also provided with inlet 3, supplied by pressure pump 4, through which an olefinic gas, or mixture of such gas with any other gas desired, may be supplied under any convenient pressure. As shown, the vessel is particularly adapted for electrolytic purposes and may or may not be provided with a diaphragm, shown in dotted line as 5. Such a diaphragm may be of any usual material and divide the cell into an anode chamber A and cathode chamber C. This diaphragm is advantageously perforated above the liquid level as at 6 to allow equalization of gas pressure on the two sides. This diaphragm may be, and in the present specific modification of my invention advantageously is, omitted. The vessel is provided with cover 7 which may be clamped on in any suitable manner. This cover carries hood 8 having communication with the chamber of the vessel through orifice 9. This equalizes the chamber pressure with that of the hood 8. Within this hood is mounted any simple type of blower or pump, shown here as rotary blower 10. This blower has communication with the gas space within the chamber at 11 and is provided with conduit 12 extending nearly to the bottom of the vessel. The lid may also be provided with two conduits 13 and 14 placed on each side of the diaphragm and valved respectively at 15 and 16. These two conduits unite as conduit 17, valved at 18, which expands into chamber 19. Above this chamber is a length of piping 20, valved at 21, and connecting with another chamber 22, above which is valve 23 and hopper or funnel 24. Equalizing pipe 25, valved at 26, allows equalization of pressure. This last described device enables equalization of gas pressure with that of the chamber by opening appropriate valves; and it also allows liquid to be fed to either side of the diaphragm and without disturbance of the chamber pressure. For example, a liquid may be fed into 24, and, valves 26 and 21 being closed, may be fed into 22 by opening valve 23. By now closing valve 23 and opening 26 and 21, (valve 18 being normally open) the liquid may be caused to pass down into 19 and thence by 16 or 15, the liquid may be discharged on either side of the diphragm.

Anode 27 and cathode 28 may be of any suitable metal or structure adapted to a particular electrolysis.

In one embodiment of the present invention using electrolytic oxidation the diaphragm 5 is omitted, allowing free communication between the liquids in the two compartments. The vessel is charged with a solution of sodium chlorid of such strength as to give good conduction. Ethylene, or gas containing ethylene, may now be introduced into the apparatus by means of pump 4 at any pressure desired, say, 4 or 5 atmospheres. Upon now starting blower 10 into operation, the liquid is kept thoroughly saturated with dissolved ethylene. It is advantageous to run this pump or blower at such a rate that the liquid is maintained as more or less of an emulsion of gas and liquid; as a foamy mass. Upon now passing a current through the device in such a manner that 27 becomes the anode, the ethylene is oxidized to glycol which goes into solution. The oxidizing influence developed at the anode whether this be due to nascent chlorin or oxygen or oxidation products of chlorin or of the chlorid, oxidizes the dissolved olefins. This oxidation may be aided by the presence of catalysts which superadd a function of absorbing gas and redelivering to the liquid. In the present embodiment of my invention I use platinum group metals as this catalyst. Platinum black or colloidal palladium may be used.

It is best to limit the rate of development of the oxidizing action at the anode according to the rate of solution of the ethylene so that an excess of ethylene shall remain in the solution at all times where a continuous operation is used. This is for the reason that should the solution temporarily become exhausted of ethylene while oxidation is still being developed there is the danger of oxidation of glycol to form glycolic acid, etc. For this reason in continuous operation the current should be regulated in correspondence with the supply of dissolved ethylene. The apparatus may be run discontinuously, a charge of ethylene, or gas containing the same, as for example, a pyrolytic oil gas containing say 40 or 50 per cent. of ethylene and other olefins, being introduced and the pump 10 being kept in operation until the ethylene of the charge has become absorbed and oxidized. At this time the current may be interrupted and waste gas vented through 14. Another charge of ethylene, or gas containing the same, may now be introduced and so on until the concentration of glycol in the solution becomes as high as may be desired. Where no diaphragm is used there is no substantial change in the solution as regards alkalinity or acidity. If a neutral salt solution is used at the beginning it is neutral at the end. Where the supply of ethylene is sufficient and the catalyst is of suitable character there will be no oxidized chlorin compounds at the end of the operation. The final result therefore will be a neutral saline solution containing glycol; and from this solution glycol may be isolated by ways similar to the ways of recovering glycerin from salt lyes, its properties, as well as its uses, being very similar to those of glycerin. Like glycerin it is a dense, high boiling liquid which however can be blown over readily with steam. The anode may be of platinum, carbon or lead. The fluid surrounding the anode may contain, in addition to the catalyst, a carrier oxydant, such as a chromium salt, a cerium salt, a manganese salt, etc. A little platinum black, palladium or the like may be kept suspended in the liquid. Other bodies having an absorbent action but not so much catalytic action, such as charcoal and bone-black may be used. But I regard the platinum group metals, which have both absorbent and catalytic properties, as the best for the present purposes.

Using salt solution as the electrolyte, the cell may be worked under conditions analogous to those used in making hypochlorites. In such an event, the cathode should be of small effective area to give a high current density and permit escape of hydrogen without reduction of oxidized chlorin compounds. It is desirable to keep the temperatures low, ordinary temperature being suitable, and to maintain an excess of ethylene dissolved in the solution.

Operating with pure ethylene, such as that made by the hydrogenation of acetylene, in the cell, pure glycol will be formed. But with pyrolytic gases containing ethylene, such as those obtained by cracking oils at low temperatures, there will usually be propylene and the butylenes present. These also make valuable glycols. The present process may be used in connection with the pure propylene or the pure butylenes in making propylene glycol and butylene glycols.

Apparatus of the type described and shown may also be used for non-electrolytic methods under the present invention. By omitting diaphragm 5, anode 27 and cathode 28, an oxidizing liquid may be fed in at a regulated rate through funnel 24 and connected parts. This oxidizing liquid may be a milk of bleaching powder. It contains hypochlorite of calcium which may be regarded as an oxidation product of calcium chlorid. It may be made in an electrolytic cell in the same way as is described for sodium hypochlorite but ordinarily is made by producing chlorin in a suitable cell and contacting it with lime elsewhere. Bleaching powder and water may be fed in through 24 until the apparatus is charged to a sufficient extent. In this charge may be a very small proportion of a cobalt salt. Ethylene may then be fed in and circulated as before. In the presence of a cobalt salt as a catalyst hypochlorite solutions, such as those made from bleaching powder, give up oxygen at a slow rate and this oxygen oxidizes the ethylene. The rate may be controlled by controlling the amount of cobalt salt or other catalyst present and should be such that the rate of development of oxygen does not exceed the rate of solution of ethylene.

The apparatus may be maintained at any suitable pressure and temperature. Ethylene or a mixture of ethylene and air may be fed in and circulated. The liquid may be kept charged with suspended or dissolved catalytics, such as platinum black, palladium, alkaline manganates, etc. The ethylene in solution should always be in excess to cause oxidation to affect it rather than its oxidation products.

In using weak ethylenic gases, such as those from cracking oil, from acetylene hydrogenated by nickel in the presence of a diluent gas, etc., if operating with a single electrolytic cell discontinuously, that is, with successive charges of gas introduced and circulated until the ethylene is absorbed, there is a periodic variation in the strength of current required. For example, with a fresh charge of oil gas containing, say, 45 per cent. ethylene and other olefins with the

residue largely ethane, the absorption of the gas is at first relatively rapid, but as the ethylene disappears solution becomes slower; being for instance considerably slower when the richness in ethylene is reduced to 30 per cent. This may need a corresponding change in the current supply. By operating a number of cells in series, as regards the gas feed, each such cell venting gas into the next in series, this inconvenience may be overcome, each cell then operating with gases of a constant average content; the first, say with gas averaging 40 per cent., the next with gas averaging 30 per cent. and so on, each cell then having a constant current supply.

Gas of any desired degree of purity may however be obtained from lean ethylenic gas by scrubbing the latter by a countercurrent of spirits of turpentine, heavy oil or any other solvent for ethylene, exhausting the charged solvent by vacuum or heat, or both, and returning the exhausted solvent for fresh use with the gas to be scrubbed. The rich ethylene obtained by exhausting the charged oil may then be oxidized.

In the specific embodiment of my invention claimed in the present case the liquid forming the electrolyte is a solution of common salt and electrolysis is in an undivided cell. In this liquid oxidation with or without development of oxidized chlorids or oxidized chlorin compounds occurs. The liquid is kept saturated with ethylene or another olefin; or with the mixed olefins of oil gas and absorption and oxidation are aided by the presence of an absorbent and catalytic body which is best one of the platinum group metals, such as platinum or palladium, though other catalysts may be used. The oxidized chlorid or oxidized compound of chlorin may however be produced elsewhere, and supplied to a separate reaction chamber and there treated with the olefin. Whether the oxidized chlorin compound be developed, so to speak, *in situ*, or whether it be supplied from another source, the solution however should be kept as nearly as may be saturated with dissolved ethylene or olefins during the action. In the absence of an excess of olefin, such carrier catalysts or oxidizing catalysts as platinum black, palladium, etc., may carry the oxidation further than is here desired.

What I claim is:—

1. The process of making glycols which comprises electrolyzing a chlorid solution in a diaphragmless cell in such manner as to oxidize said chlorid, and maintaining the electrolyte saturated with a gaseous olefiin in the presence of an oxygen carrying catalyst.

2. The process of making glycols which comprises electrolyzing sodium chlorid in a diaphragmless cell in such manner as to oxidize said chlorid, and maintaining the electrolyte saturated with a gaseous olefin in the presence of an oxygen carrying catalyst.

3. The process of making glycols which comprises electrolyzing a chlorid solution in a diaphragmless cell in such manner as to oxidize said chlorid, and maintaining the electrolyte saturated with oil gas in the presence of a platinum group metal as a catalyst.

4. The process of making glycols which comprises electrolyzing sodium chlorid in a diaphragmless cell in such manner as to oxidize said chlorid, and maintaining the electrolyte saturated with a gaseous olefin in the presence of a platinum group metal as a catalyst.

5. The process of making glycols which comprises maintaining a solution of an oxidized chlorid salt saturated with a dissolved olefin in the presence of a platinum group metal as a catalyst.

6. The process of making glycols which comprises maintaining a solution of an oxidized chlorid salt saturated with oil gas in the presence of a catalyst.

7. The process of making glycols which comprises maintaining a solution of an oxidized chlorid salt saturated with oil gas in the presence of a platinum group metal as a catalyst.

8. The process of making glycols which comprises agitating a solution containing a dissolved oxidized chlorin salt with oil gas in the presence of a catalyst.

9. The process of making glycols which comprises agitating a solution containing a dissolved oxidized chlorin salt with oil gas in the presence of a platinum group metal as a catalyst.

10. The process of oxidizing gaseous olefins which comprises emulsifying such olefins with a chlorid electrolyte during electrolysis and in the presence of a catalyst.

11. The process of oxidizing gaseous olefins which comprises emulsifying such olefins with a liquid containing products of the electrolytic oxidation of a chlorid and also containing a platinum group metal as a catalyst.

12. The process of oxidizing gaseous olefins which comprises emulsifying such olefins with a liquid containing a salt of an oxygenated acid of chlorin and also containing a platinum group metal as a catalyst.

13. The process of oxidizing gaseous olefins which comprises electrolyzing a chlorid electrolyte in a diaphragmless cell and emulsifying the electrolyte with such olefins in the presence of a platinum group metal as a catalyst.

14. The process of making useful products from oil gas which comprises emulsifying oil gas with a chlorid electrolyte during electrolysis and in the presence of a catalyst.

15. The process of oxidizing gaseous olefins which comprises electrolyzing a chlorid electrolyte in a diaphragmless cell and emulsifying the electrolyte with oil gas in the presence of a platinum group metal as a catalyst.

16. The process of making glycols, which comprises bringing an olefin in gaseous state in contact with a solution of an oxidized halid salt in the presence of a platinum group metal as a catalyst.

17. In the process of producing oxidation products of the olefins, the step which consists in subjecting an olefin to the action of an oxidizing agent in the presence of a catalyst while maintaining the olefin in excess.

In testimony whereof, I affix my signature.

K. P. McELROY.

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CATALYSIS AND CATALYTIC AGENTS IN CHEMICAL PROCESSES.

BY

HUGH S. TAYLOR.

* * *

Each class of reaction has its own class of catalytic agent and each of these classes may similarly be circumscribed as to members. Thus, for example, it may be seen that as catalysts for oxidation processes, oxides are an important class of catalysts. Of the oxide class, however, it is remarkable that only a limited number function at all prominently as catalysts. The oxides of well-defined stability are conspicuously absent. The oxides of the alkali metals, to a less extent those of the alkaline earths, definite irreducible oxides such as magnesium oxide and aluminium oxide are not frequently to be found among the oxide catalysts of oxidation. On the other hand, the labile oxides such as those of iron, copper, manganese, silver, vanadium, tungsten, molybdenum uranium, chronium, cobalt and cerium are frequently to be noted in a systematic study of such oxide catalysts of oxidation. * * *